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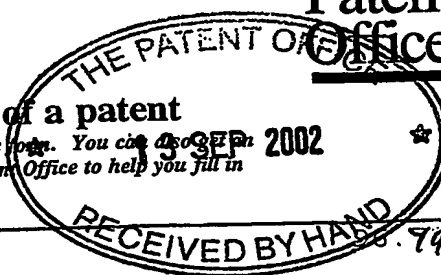
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P.O. Box 6034
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N-5892 Bergan
Norway

08083867001

Patents ADP number (if you know it)

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14SEP02 E748211-1 000027
P01/7700 0.00-0221304.9

0221304.9

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Co-Production of Hydrogen

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Frank B. Dehn & Co.

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|  | FUTURE ENERGY PLANTS Technical Summary Report M1 1 - INTRODUCTION | Doc.no. RP_21205_PR_01 |
| Project: ZEG | | Issue: A Date: 09.09.2002 |
| | | Page: 5 of 73 |

1 INTRODUCTION

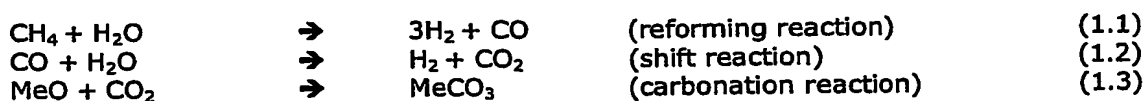
The Zero Emission Gas Power project (ZEG) was launched in 2001 by the Institute for Energy Technology (IFE), Christian Michelsen Research (CMR) and Prototech. The overall objective is to develop technology facilitating energy efficient combined production of electric power and hydrogen from natural gas, with integrated capture of CO₂. The captured CO₂ can then either be sequestered or used in other industrial processes. The ZEG project does not address the problem of CO₂ sequestration as such. The duration of the project is scheduled to 4.0 years with a final phase comprising a laboratory demonstration of a system producing electricity and hydrogen from natural gas and where CO₂ separation is included. The project is part of the KLIMATEK program funded by the Norwegian Research Council (NFR).

The general idea behind the project is to establish technology that makes it possible to build gas stations supplying hydrogen to vehicles and electricity to the local community. Furthermore, the technology should also be scalable to allow realization of large¹ zero emission gas power stations, as well. The latter may be of particular importance, when pursuing cost effective and safe solutions to the CO₂-storage problem.

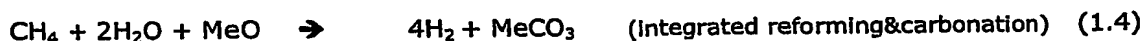
The background for the project is the need to develop technology to deal with the problem that we for the foreseeable future will rely on use of fossil fuels whilst the requirements for reduced environmental impacts from production of electric power and energy use, are expected to increase significantly. In addition, e.g. new energy conversions processes with reduced environmental impacts, will still have to be cost effective and in particular the energy efficiency must be improved compared to current technology.

An important aspect of the ZEG project is that environmental friendly power generation from natural gas combined with production of hydrogen address both the global warming problem and the problem of local pollution from the transportation sector.

Initially, the basic concept to meet the above objective, was based on combining a high temperature solid oxide fuel cell (SOFC) system for generating electric power, with a novel natural gas reforming reaction for simultaneous production of hydrogen and capture of CO₂. The reforming process can be described as follows




resulting in the following overall reaction



where MeO is a metal oxide. Utilizing heat from the SOFC system, CO₂ is then desorbed from the carbonate according to



¹ 100 MW range

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| prototech  | FUTURE ENERGY PLANTS Technical Summary Report M1 1 - INTRODUCTION | Doc.no. RP_21205_PR_01 Issue: A Date: 09.09.2002 |
| Project: ZEG | | Page: 6 of 73 |

and the metal oxide is then recycled in the process. If calcium oxide (CaO) is used eq.(1.5) is referred to as the calcination reaction and eq.(1.4) is almost thermally neutral.

The carbonation/calcination process matches the reforming/electrochemical conversion process of the fuel cell system:

- the exothermic carbonation reaction is coupled to the endothermic reforming reaction
- the equilibrium of the overall reaction [R7, R8] gives at least 95% hydrogen (dry basis) at standard reforming temperatures (approximately 500°C)
- the exothermic electrochemical reaction is coupled to the endothermic calcination reaction
- the calcination reaction is compatible with the high operating temperature of the SOFC

An alternative concept to the above process, would be to combine an SOFC system with a hydrogen permeable membrane integrated in the SOFC prereformer, in order to produce hydrogen.

In both concepts, as described above, the high fuel-to-electrical-power efficiency and the high temperature waste heat, characterizing SOFC systems, are utilized. Furthermore, the fact that fuel and air streams are separate through the SOFC system, is vital to achieve energy efficient separation of CO₂ from the electrical power generation process.

The main technical responsibilities in the project are divided between the partners according to

- IFE : Combined reforming and carbonation reactions, calcination reaction and reactor technology.
- Prototech : SOFC technology and system design.
- CMR : Supporting technologies.

This report is part of the documentation prepared for Milestone 1 in the project, originally planned for December 2002. The Milestone 1 documentation was originally planned to comprise a review of the initial experimental and technological results as well as necessary adjustments to the system concepts. Furthermore, preliminary mass- and energy-balance for the system concepts considered, should be included.

In combination with [R8], [R10], [R11], [R12], the present report comprise a summary of the work carried out in the period September 2001 until September 2002.


This report focuses on system analysis of four process concepts in combination with a review of the technological basis for the processes. Chapter 3 gives a summary of the report. Chapter 4 describes four different system configurations that have been analysed with respect to system performance, costs and technological risks. Chapter 5 presents details about the system analyses. Results from these analyses with respect to efficiencies and output are given in chapter 6.

Cost estimations for the plant, cost of electricity and cost of hydrogen are included in chapter 7. Detailed listing of temperatures, mass and volume flows used for the cost analyses are given in Appendix A, while details on the cost analyses are given in Appendix B.

Unconventional technologies introduced in the concepts include integrated reforming&carbonation and calcination, membrane technologies and oxygen pumping. Sizing and costing of process steps including these technologies are based on laboratory experiments in combination with cost data for similar processes. The technical risks are discussed in chapter 8. The theoretical and experimental work related to the new technologies is summarized in Chapter 9, while a more detailed presentation is given in Appendices C, D and E.

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| Project: ZEG | | Issue: A Date: 09.09.2002 Page: 7 of 73 |


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2.1 References

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<http://www.entek.chalmers.se/~anly/symp/testsymp2001.htm>.
- [R2] **SECA FUEL CELLS FOR LARGER APPLICATIONS**, Jan Thijssen, ARTHUR D.LITTLE, 3rd Annual SECA Workshop, Washington, DC, March 21-22, 2002.
- [R3] **A COMPARATIVE ANALYSIS OF IGCCs WITH CO₂ SEQUESTRATION**, Paolo Chiesa, Stefano Consonni, Giovanni Lozza - Dipartimento di Energetica, Politecnico di Milano, P.zza Leonardo da Vinci, 32 - 20133 Milan - ITALY, 1999.
- [R4] **A PORTFOLIO SELECTION APPROACH FOR POWER PLANT CO₂ CAPTURE, SEPARATION AND R&D OPTIONS**, Dale Simbeck, Vice President Technology - SFA Pacific, Inc., Mountain View, CA, USA, 1999.
- [R5] **SURVEY OF ECONOMICS OF HYDROGEN TECHNOLOGIES**, C.E.G. Padró and V. Putsche, NREL, NREL/TP-570-27079, National Renewable Energy Laboratory, 1617 Cole Boulevard, Golden, Colorado 80401-3393, September 1999.
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- [R7] **HYDROGEN PRODUCTION BY STEAM REFORMING OF METHANE IN THE PRESENCE OF A CO₂ ACCEPTOR**, A. Lopez Ortiz, B. Balasubramanian, and Douglas P. Harrison, Gordon A. and Mary Cain Department of Chemical Engineering, Louisiana State University, Baton Rouge, LA 70803 USA.
- [R8] **FUTURE ENERGY PLANTS - SUBPROJECT 1 - INTEGRATED REFORMING AND CO₂-CAPTURE. TECHNICAL REPORT, MILESTONE**. IFE/KR/F-2002/123, Meyer J., Eriksen D.Ø., Glöckner R. and Ørjasæter B. Institute for Energy Technology, Kjeller, Norway, September 2002.
- [R9] **ASSESSMENT OF MEMBRANE TECHNOLOGY FOR CARBON DIOXIDE REMOVAL IN POWER PLANTS**, Author P.H.M. Feron, TNO Institute of Environmental and Energy Technology, September 1992.
- [R10] **FUTURE ENERGY STATIONS - A FEASIBILITY STUDY FOR SENSORS AND CONTROL TECHNOLOGIES**, Kjell Elvind Frøysa and Ole Brix. CMR-02-F10014, Christian Michelsen Research, Bergen, Norway, August 2002.
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| prototech  | FUTURE ENERGY PLANTS Technical Summary Report M1 2 - REFERENCES AND DEFINITIONS | Doc.no. RP_21205_PR_01 Issue: A Date: 09.09.2002 Page: 8 of 73 |
| Project: ZEG | | |

[R12] **FUTURE ENERGY PLANTS; CO-PRODUCTION OF ELECTRICAL POWER AND HYDROGEN FROM NATURAL GAS WITH INTEGRATED CO₂ CAPTURE, TECHNICAL SUMMARY REPORT, IFE/KR/F-2002/130.**

[R13] **OVERVIEW OF THE ZECA (ZERO EMISSION COAL ALLIANCE) TECHNOLOGY, LA-UR-00-6002, Hans-Joachim Ziock, Klaus S. Lackner, Los Alamos National Laboratory, Los Alamos, NM 87545.**

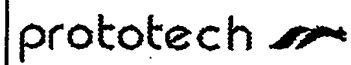
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2.2 Definitions

| | |
|----------------------------------|---|
| A_m | Membrane Area |
| Am | Membrane Area |
| Approx. | Approximately |
| BOP | Balance Of Plant |
| CC | Combined cycle |
| COE | Cost of Electricity |
| COH ₂ | Cost of Hydrogen |
| CV | Cell Voltage |
| d | Day(s) |
| DC | Direct Current |
| Dry basis | Dry atmosphere (water/steam not present) |
| DEA | Aqueous Solution of Diethanolamine |
| ΔT | Temperature difference |
| Eff | Efficiency |
| EI | Electrical/Electric/Electricity (depending on the context) |
| ϕ | Molar Flux [mol/(m ² sec)] |
| Φ | Molar Flux [mol/(m ² sec)] |
| F | Faraday constant |
| FB | Fluidised Bed |
| FC | Fuel Cell(s) |
| FU | Fuel Utilisation |
| GJ _{H2} | Throughput of Hydrogen [GJ] produced in one year |
| GT | Gas Turbine |
| (g) | gas |
| h | Hour(s) |
| hr | Hour(s) |
| HV | Heating Value |
| HHV | Higher Heating Value |
| I | Interest rate |
| Integrated Reforming&Carbonation | Natural Gas-water reforming reaction integrated with the CO shift reaction and a carbonation reaction ($CH_4 + H_2O = 3H_2 + CO$ and $CO + H_2O = H_2 + CO_2$ and $MeO + CO_2 = MeCO_3$ integrated to give the overall reaction $CH_4 + 2H_2O + MeO = 4H_2 + MeCO_3$); MeO can be for instance CaO. |
| K_p | Reaction's equilibrium constant |
| K-type thermocouple | Device to measure temperature |
| LCF=load capacity factor | Ratio between the hours corresponding to full load operation in one year and 8760 (where $8760[hr] = 365[d] * 24[hr/d]$) |
| LHV | Lower Heating Value |
| Log | Base 10 logarithmic function |
| MeO | Metal Oxide |
| Met-oxide | Metal Oxide |

¹ Natural Gas is considered for explanation simplicity to be 100% methane

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|  | FUTURE ENERGY PLANTS Technical Summary Report M1 2 - REFERENCES AND DEFINITIONS | Doc.no. RP_21205_PR_01 Issue: A Date: 09.09.2002 |
| Project: ZEG | | Page: 10 of 73 |

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|----------------------|--|
| mill\$ | 10^{-3} [\$] = 10^{-3} USD |
| M\$ | 10^6 [\$] = 10^6 USD |
| MW _{el} | [MW] electric |
| MW _{th} | [MW] thermal |
| N | Normal |
| NG | Natural Gas |
| NGCC | Natural Gas Combined Cycle |
| NGCC _{GT} | Natural Gas Combined Cycle, Gas Turbine based |
| OD | Outer Diameter |
| O&M | Operating and Maintenance |
| Oxygen Pump | electrochemical device forcing oxygen ions through an electrolyte by application of external voltage between electrodes on each side of the electrolyte |
| P | Total pressure |
| Permeate gas(es) | Gas(es) passing through a membrane |
| Pd-membrane | Palladium Membrane |
| P _f | Feed Pressure |
| P _f | Feed Pressure |
| P _p | Permeate Pressure (Pressure of permeate gas(es)) |
| P _p | Permeate Pressure (Pressure of permeate gas(es)) |
| P _{xx} | Partial pressure of component xx |
| Recycle ratio | Ratio between the mass flow of the recycled stream and the mass flow of the incoming stream |
| Reforming+shift | Reforming and Shift coupled reactions (CH ₄ +H ₂ O=3H ₂ +CO and CO+H ₂ O=H ₂ +CO ₂ coupled to give CH ₄ +2H ₂ O=4H ₂ +CO ₂) |
| Retentate gas(es) | Gas(es) <u>not</u> passing through a membrane |
| S | Size |
| SOFC | Solid Oxide Fuel Cell |
| Stack(s) | Structural and operational Unit in a fuel cell system, made of a number of individual cells |
| Tech | Technology |
| T _m | Absolute Temperature of the membrane [K] |
| T _m | Absolute Temperature of the membrane [K] |
| t _m | Thickness of the Membrane |
| tm | Thickness of the Membrane |
| USD | United States Dollar [\$] |
| Volume _{pd} | Volume of Palladium |

Unless otherwise specified calculations and values of the present report are LHV based.

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3 SUMMARY

Four different system concepts for electricity and hydrogen production with inherent CO₂ capture using an integrated SOFC power plant have been developed. The characteristics of the different systems are:

- ZEG 1** SOFC power plant with afterburner using pure oxygen from an oxygen pump, thermally integrated with a hydrogen production unit including an integrated reforming&carbonation unit and a calcination reactor for CO₂ capture. One CO₂ stream comes from the SOFC stacks exhaust, and one from the calcination reactor.
- ZEG 2** A common integrated reforming&carbonation unit generates hydrogen both for the SOFC power plant and the hydrogen production. The afterburner of the SOFC stacks is fed with air, as the anode exhaust stream contains no CO₂. The system also includes a calcination reactor for CO₂ capture. All the CO₂ comes from the calcination reactor.
- ZEG 3** SOFC power plant with afterburner using pure oxygen from an oxygen pump. The NG reforming takes place in a standard natural gas reformer, and an integrated H₂ separation membrane separates H₂ for hydrogen production. All the CO₂ comes from the SOFC stacks exhaust.
- ZEG 4** SOFC power plant operated on high throughflow of hydrogen in a hydrogen recycle loop. Water is continuously removed from the recycle loop. The NG is reformed in an integrated reforming&carbonation unit producing hydrogen both for the SOFC power plant and for hydrogen production. The system also includes a calcination reactor for CO₂ capture. All the CO₂ comes from the calcination reactor. The system does not include an afterburner, as the system fuel utilisation is 100%. The system has a theoretical electrical and a total efficiency approaching 100%, only limited by BOP losses and is identified as a very interesting and novel concept.

Process simulations have been performed for all the four concepts, and the output includes temperatures, mass and volume flows, compositions, heat balances as well as efficiencies. A summary of the efficiencies is given in figure 3-1.

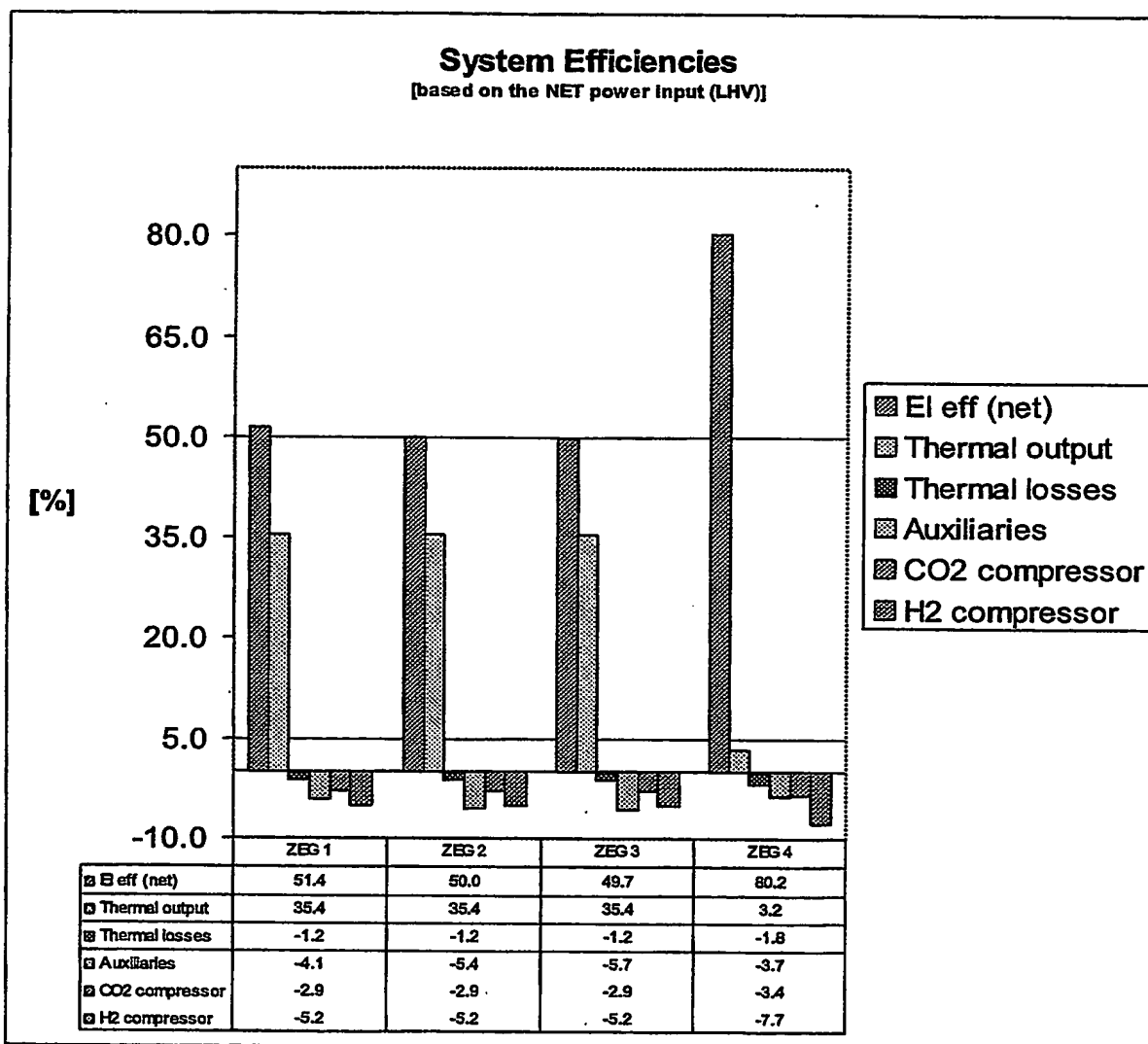


Figure 3-1 System efficiencies. $\text{NET_Power_Input} = \text{TOT_Power_Input} - \text{H}_2\text{_Output}$ where: TOT_Power_Input is the total power input LHV based $[\text{MW}_{\text{th}}]$ of the fuel entering the system; $\text{H}_2\text{_Output}$ is the power output LHV based $[\text{MW}_{\text{th}}]$ of the H_2 leaving the system; $\text{Net_electric_output}$ is the gross electric output of the system minus the electric power required to drive the oxygen pump and the air, CO_2 , H_2 compressors. By not compressing the H_2 , for example, the *El eff (net)* would increase 5.2% for ZEG 1, 2, 3 and 7.7% for ZEG 4.

The cost of electricity and the cost of hydrogen have been analysed based on investment cost and operating costs for all major cost elements. Sizing and costing of process steps including the new technologies are based on laboratory experiments in combination with cost data for similar processes.

The results show that cost of electricity may be significantly lower in ZEG 4 than for one of the assumed most efficient comparable system based on thermodynamic cycles (NGCC_{GT} with CO₂ scrubbing). The results also show that hydrogen production with integrated CO₂ capture may be performed more cost efficiently in the ZEG concepts than by conventional Steam Methane Reforming without CO₂ capture (the evaluation of the ZEG concepts is based on a Small Size Facility outline).

A summary of the cost analysis is given in figures 3-2 and 3-3.

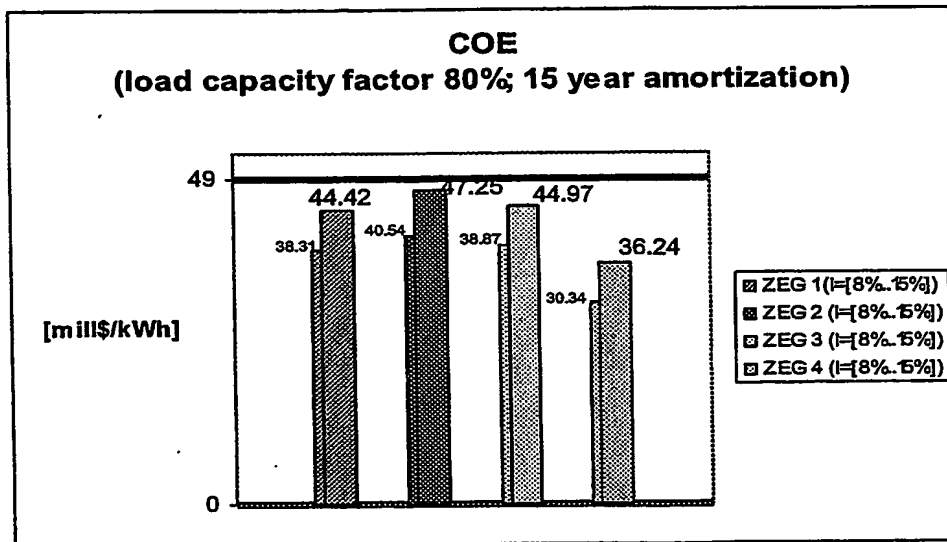


Figure 3-2 Estimated COE; for each of the ZEG concepts the interval of COE variation is showed according to the interest varying in the interval [8%..15%]; 49 [mill\$/kWh] indicates the typical value for NGCC ("H") + flue gas CO₂ scrubber (I=15%) [R4]

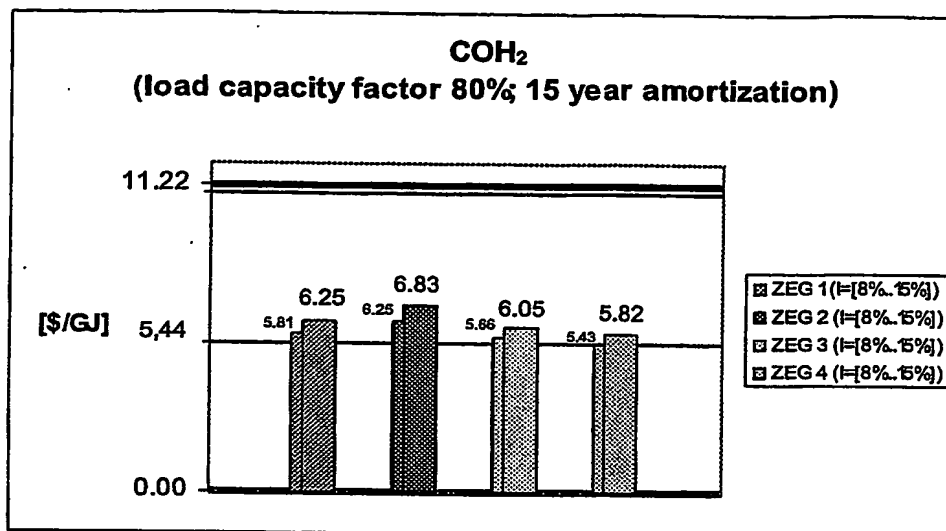



Figure 3-3 Estimated COH₂; for each of the ZEG concepts the interval of COH₂ variation is showed according to the interest varying in the interval [8%..15%]; 5.44 [\$/GJ] indicates the typical lowest limit for Large Size Facilities Steam Methane Reforming (SMR) technology [R5]; 11.22 [\$/GJ] indicates the typical value for Small Size Facilities (i.e. ≤0.27 million [Nm³/day]) Steam Methane Reforming (SMR) technology [R5]

The system concepts and included technologies have been analysed with respect to technical risks, and shows that the ZEG 4 concept, identified as the most promising concept has the highest technological risk. The technical risk of this concept is mainly related to the close thermal integration required to obtain the very high efficiency, however even with a not optimised thermal integration this system will outperform the others.

A significant theoretical and experimental work has been performed to gather knowledge about the reforming&carbonation and calcination processes. Numerous experiments have also been performed to characterise the calcination and the carbonation processes. The experiments show that the reactions rates are sufficient for commercial application of the processes. Although some degradation in the absorbing capacity of the CaO is observed, the implementation of the processes still seem feasible. However, further work should aim on improving the long term stability of the absorbent. Fixed bed plate heat-exchangers are used as a baseline in the cost analysis and in the heat transfer analysis. Experiments show that this concept is feasible. Experiments have been performed both at IFE and Prototech. Prototech has so far focused on experiments generating data for the implementation of the baseline concept, while IFE is performing a broader range of experiments to support evaluation of alternative reactor designs. However, different reactor concepts have also been evaluated. Further work is required to make a final choice of reactor concept.

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|  | FUTURE ENERGY PLANTS Technical Summary Report M1 | Doc.no. RP_21205_PR_01 Issue: A Date: 09.09.2002 |
| Project: ZEG | 3 - SUMMARY | Page: 15 of 73 |

Unconventional technologies introduced in the concepts include integrated reforming&carbonation and calcination, membrane technologies and oxygen pumping, and significant theoretical work and laboratory experiments have been performed to support the design, process simulations and cost analysis of the different concepts.

As a part of the extensive SOFC development work carried out at Prototech since 1990, experience with manufacture and testing of oxygen pumps has been established. In addition to theoretical analysis of the oxygen pump implementation, the test set-up for the oxygen pump is under preparation.

Alternative hydrogen membrane technologies have been investigated:


- palladium membrane (Pd-membrane)
- polyimide membrane
- mixed conductor membrane

Considering the operating temperature of the catalytic reforming reactor, the best solution appears to be the use of a Pd-membrane.

The work performed so far has resulted in a new, innovative and novel concept with very high electrical efficiency. Supporting system simulations, cost analysis, theoretical and laboratory work have given increased confidence in the realisation of the concept.

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| Project: ZEG | 4 - PROCESS CONCEPTS | Page: 16 of 73 |

4 PROCESS CONCEPTS

4.1 ZEG 1 – Thermally Integrated Processes

This system consists of a stand-alone fuel cell system thermally integrated with a H₂ production system. The conceptual layout of the system is shown in figure 4-1, while the overall flow diagram is shown in figure 4-2. Temperatures, flows, compositions and heat balances are given in Appendix A.

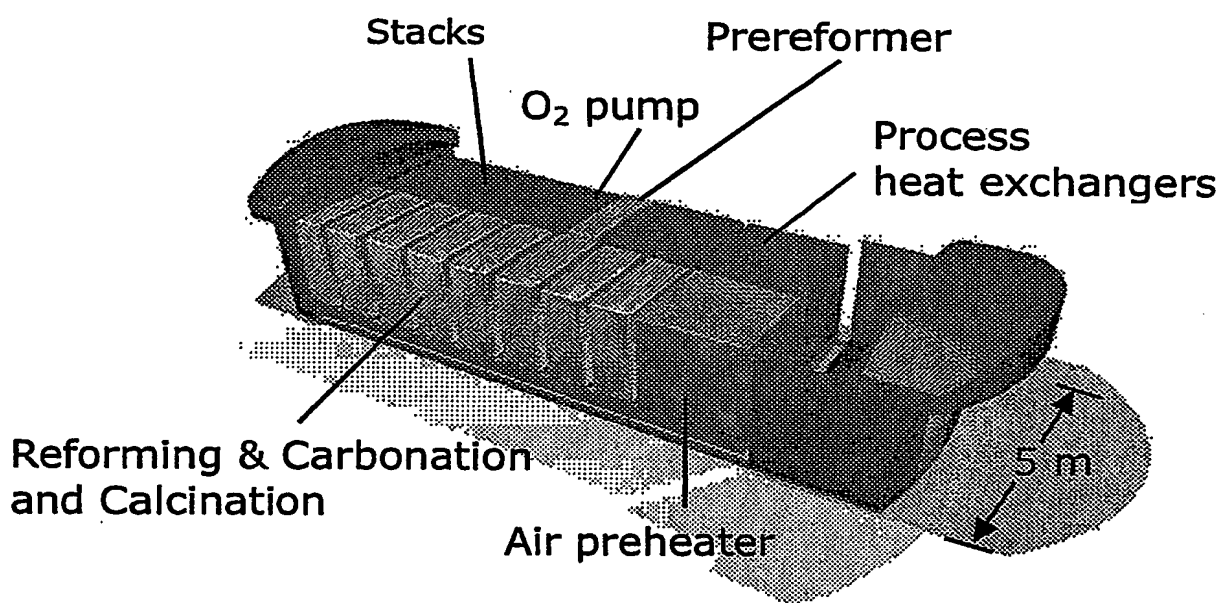


Figure 4-1 Conceptual layout of concept ZEG 1 - Thermally Integrated Processes. The shown dimensions refer to a plant with an output of 7.5 [MW_{el}] (gross DC) and 7.5 [MJ/s] of hydrogen

The basic SOFC power plant consists of a prereformer, the SOFC stacks, an afterburner to burn the residual fuel not used in the SOFC stacks, and finally heat-exchangers and a condenser to recover useful heat. The afterburner receives pure oxygen from an oxygen pump, resulting in an exhaust stream consisting of CO₂ and H₂O. On the air (cathode) side of the SOFC stacks a recycle loop is used to increase the efficiency of the system. Implementing also fuel (anode) recycle the total efficiency of the system may be further increased [R1].

The hydrogen production system consists of a steam generator, an integrated reforming&carbonation reactor, a calcination reactor (including CO₂ compression) and a hydrogen cooling&compression section. The solids may be moved continuously from the reforming&carbonation reactor to the calcination reactor and vice versa, or the reactors may change modus batchwise. The sizing and economical analyses have been performed based on a batchwise system.

In figure 4-1 is shown a feasible system layout in respect to physical dimensions of the main facilities included in the plant (SOFC stacks, oxygen pump, heat exchangers, prereformer, integrated reforming&carbonation reactors, calcination reactors). The dimensions showed on figure 4-1 (figure 4-3 for ZEG 2, figure 4-5 for ZEG 3, figure 4-7 for ZEG 4) have been derived considering the required area to heat exchange the involved thermal power in the processes (see Appendix A) and the required volume to handle the amount of solids involved in the reactors. The conceptual layout (figure 4-1) is based on a feasible batchwise project philosophy concerning the integrated reforming&carbonation and calcination reactors where the reactors alternate between being operated as reforming&carbonation reactors and calcination reactors.

The thermal integration of the SOFC power plant with the hydrogen production system transfers high temperature (950-1050°C) heat from the afterburner to the calcination reactor and low temperature heat from the SOFC stacks exhaust to the steaming&preheating section.

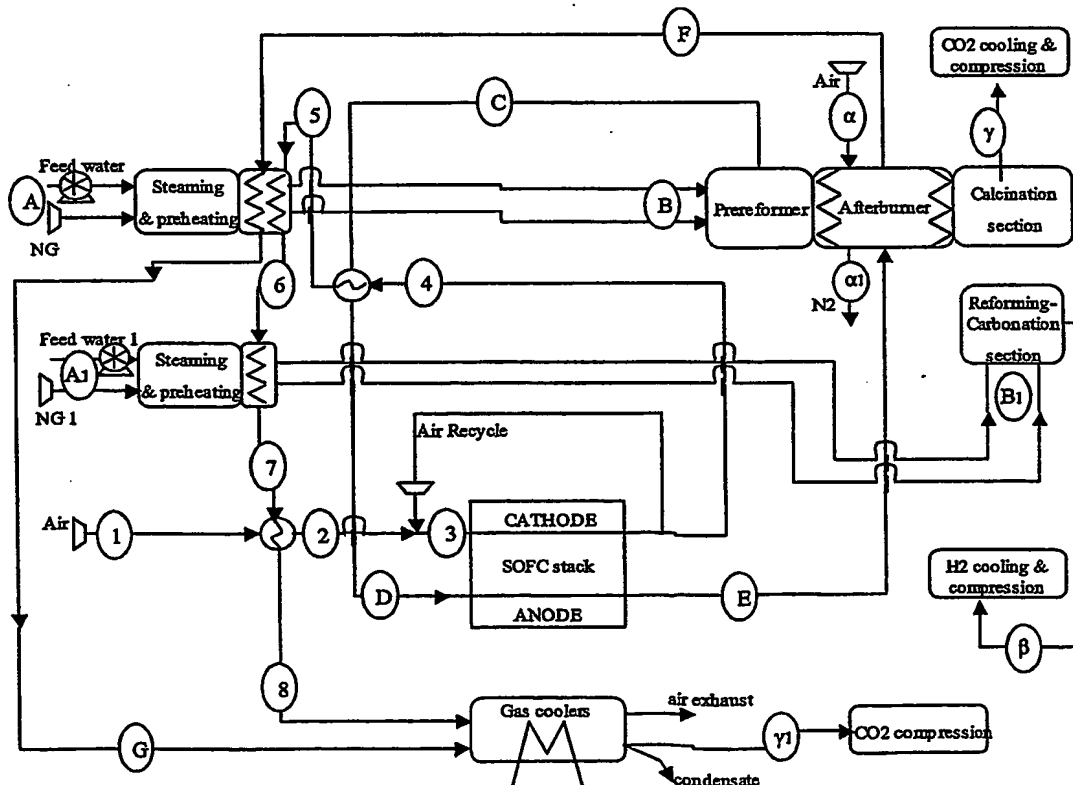


Figure 4-2 Flow diagram of ZEG 1 - Thermally Integrated Processes
 For process details see Appendix A

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4.2 ZEG 2 – Shared Reforming&Carbonation

In this concept the NG for both the SOFC stacks and the hydrogen production enters shared reforming&carbonation reactors after preheating and steam addition. A fraction of the hydrogen produced in the reforming&carbonation reactors goes to the SOFC stacks, while the remaining fraction goes to the cooling&compression section. The implications for the system compared to ZEG 1 are:

- All CO₂ is converted to carbonate, hence integrated reforming&carbonation and calcination reactors are larger
- A separate prereformer for the SOFC power plant is omitted
- The SOFC stacks are fuelled with pure H₂
- Oxygen pump is omitted, since SOFC stacks exhaust fuel consists of H₂O and H₂ and final fuel exhaust will contain only N₂ and water vapour and can be released into the atmosphere.

The system contains fewer components, but the amount of carbonate is larger than in concept ZEG 1. The conceptual layout of the system is shown in figure 4-3, while the overall flow diagram is shown in figure 4-4. Temperatures, flows, compositions and heat balances are given in Appendix A.

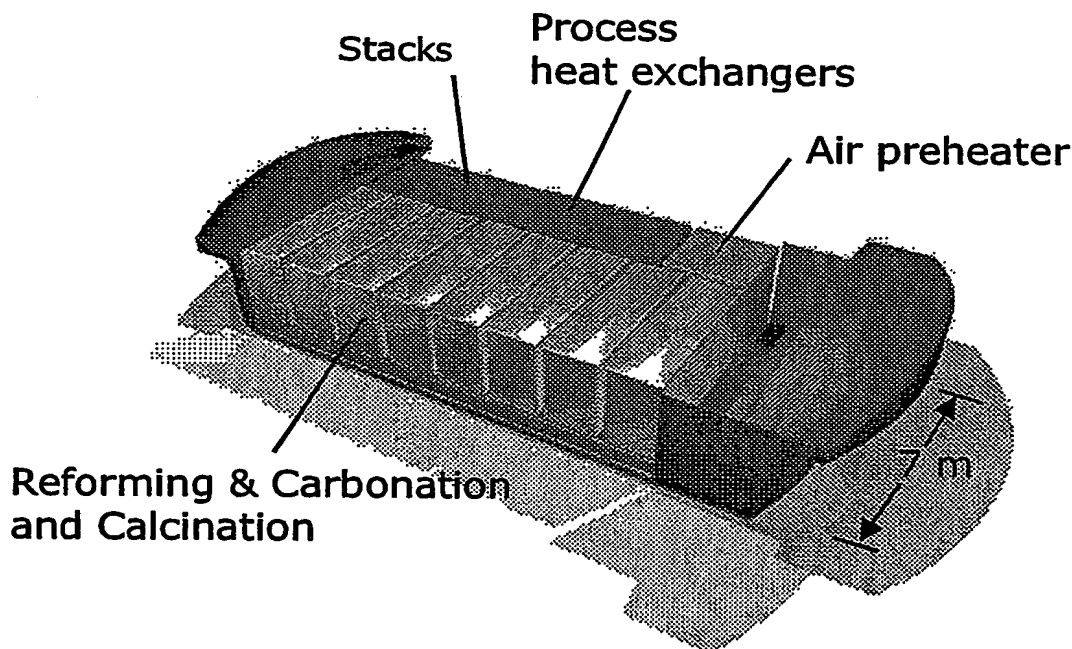


Figure 4-3 Conceptual layout of concept ZEG 2 - Shared Reforming&Carbonation. The shown dimensions refer to a plant with an output of 7.5 [MW_{el}] (gross DC) and 7.5 [MJ/s] of hydrogen

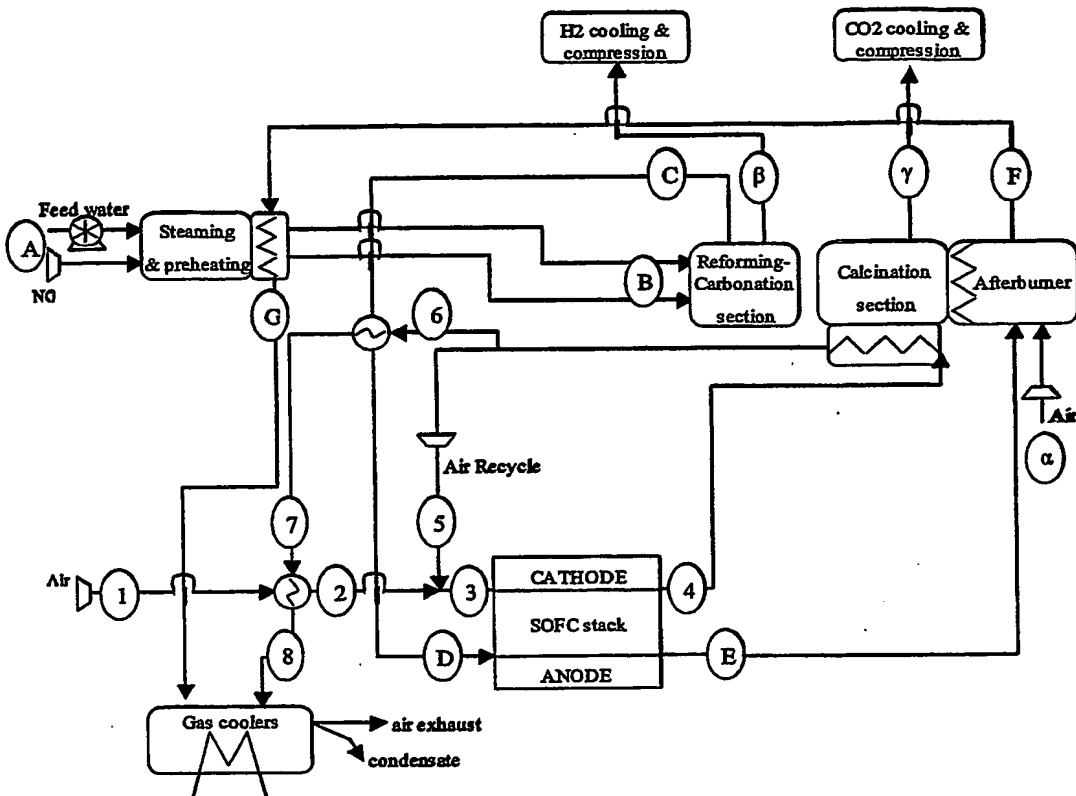



Figure 4-4 Flow diagram of ZEG 2 - Shared Reforming&Carbonation
 For process details see Appendix A

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| prototech  | FUTURE ENERGY PLANTS Technical Summary Report M1 4 - PROCESS CONCEPTS | Doc.no. RP_21205_PR_01 Issue: A Date: 09.09.2002 |
| Project: ZEG | | Page: 20 of 73 |

4.3 ZEG 3 – H₂ Membrane and Oxygen Pump

This system does not use the carbonation/calcination processes, but produces hydrogen by the use of an hydrogen membrane integrated with the reforming reactor. A fraction of the hydrogen produced in the reformer is separated by a hydrogen selective membrane, while the remaining CO rich gas is used to fuel the SOFC power plant. The SOFC can convert CO directly to electricity with practically the same efficiency as it converts hydrogen. Except for the hydrogen membrane and the hydrogen cooling&compression system, the system is identical to a basic fuel cell system. The afterburner will have to be fuelled by oxygen from an oxygen pump to produce an exhaust stream containing only H₂O and CO₂, without diluting N₂. The conceptual layout of the system is shown in figure 4-5, while the overall flow diagram is shown in figure 4-6. Temperatures, flows, compositions and heat balances are given in Appendix A.

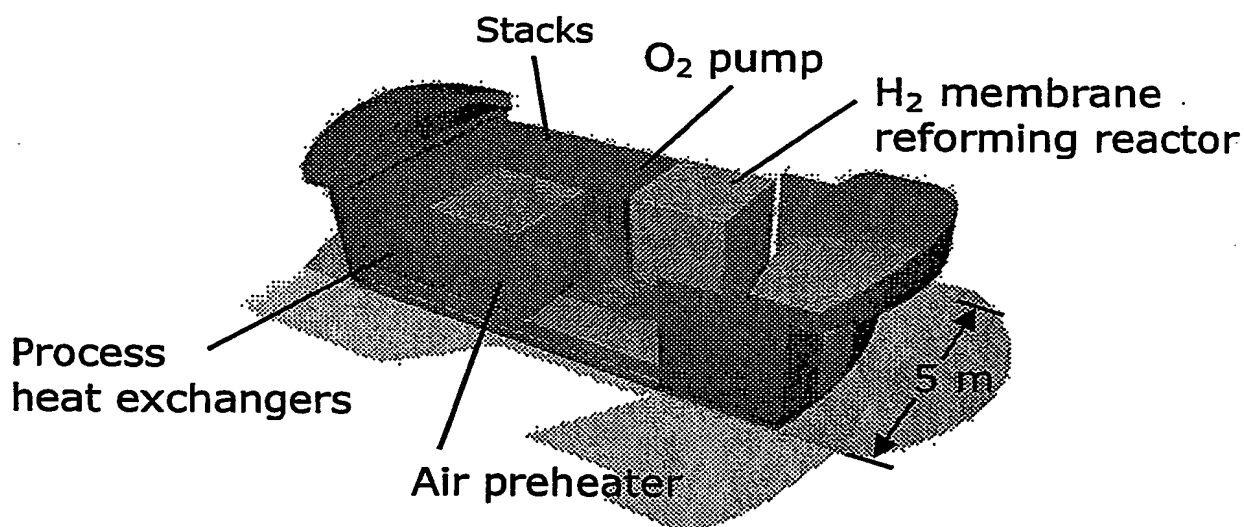


Figure 4-5 Conceptual layout of concept ZEG 3 - H₂ membrane and Oxygen Pump. The shown dimensions refer to a plant with an output of 7.5 [MW_{el}] (gross DC) and 7.5 [MJ/s] of hydrogen

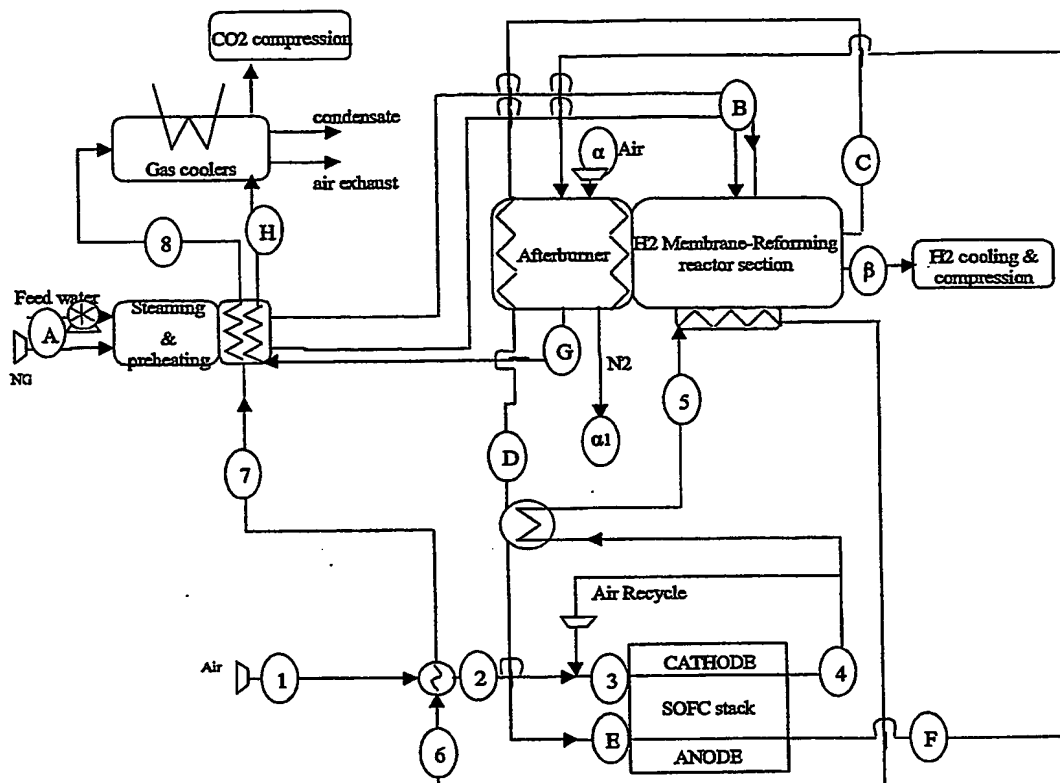



Figure 4-6 **Flow diagram of ZEG 3 - H₂ membrane and Oxygen Pump**
For process details see Appendix A

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|  | FUTURE ENERGY PLANTS Technical Summary Report M1 | Doc.no. RP_21205_PR_01 Issue: A Date: 09.09.2002 |
| Project: ZEG | 4 - PROCESS CONCEPTS | Page: 22 of 73 |

4.4 ZEG 4 – SOFC with Hydrogen Recycle

This system concept was not included in the original project proposal, but has been developed during the first phase of the project. The concept is a novel process that removes the upper limit on efficiency otherwise found in fuel cell systems and systems combining fuel cells and turbines, by further utilising the possibilities found in the integration with the integrated reforming&carbonation and calcination process. A short description of the background is included in the following.

The electrical efficiency of a system is commonly defined as

$$El_{eff} = \frac{\text{Electric_energy_output}}{\text{Energy_Input}}$$

The electric energy output of a fuel cells stack system is given by

$$\text{Electric_energy_output} = \text{Number_of_electrons} * F * \text{Cell_Voltage} * FU$$

where

| | | |
|--------------|---|---|
| F | = | Faraday constant |
| Cell_Voltage | = | the operating cell voltage of the fuel cell |
| FU | = | Fuel Utilisation of the stacks system |

In a standard fuel cell system, the fuel exhaust flows from the outlet of the fuel cell to the afterburner. The cell voltage of the fuel cell has to be lower than the Nernst potential of the fuel at the outlet, otherwise the anode is destroyed. With a fuel utilisation of 90%, the Nernst potential at the outlet of the fuel cell is 0.76V, and the fuel cell is usually run at 0.7V or lower when fuel utilisation is high.

When fuelled by CH₄ with a LHV of 802 kJ/mole and operated at a cell voltage of 0.7V, the electric energy output will be:

$$\text{Electric_energy_output} = 8 * 96487 \text{ coul / mole} * 0.7V * FU = 540 \text{ kJ / mole} * FU$$

When considering also the fuel utilisation of 90%, the net electrical efficiency of the stacks system is

$$El_{eff} = \frac{540 \text{ kJ / mole} * 90\%}{803 \text{ kJ / mole}} = 61\%$$

The ZEG 4 system concept utilises hydrogen recycle to keep the Nernst potential at the outlet of the SOFC stacks high, and this makes it possible to increase the cell voltage of the SOFC stacks without reducing the power density of the SOFC stacks. When operated at a cell voltage of 0.85V, the electric energy output will be:

$$\text{Electric_energy_output} = 8 * 96487 \text{ coul / mole} * 0.85V * FU = 656 \text{ kJ / mole} * FU$$

The water in the fuel exhaust is condensed, and the remaining hydrogen is recycled. No afterburner is then required and the fuel utilisation is 100%. This results in an electrical efficiency of

$$El_{eff} = \frac{656 \text{ kJ / mole} * 100\%}{803 \text{ kJ / mole}} = 82\%$$

It should be noted that this is not the upper limit, but a value compatible with the power output of a traditional SOFC system. When the ZEG 4 system is run at part load, the cell voltage and the electrical efficiency can be further increased.

Several systems combining a SOFC system with turbines have been developed to increase the electric efficiency of the overall power plant system. In the ZEG concepts, part of the waste heat is utilised to produce hydrogen. When combining the SOFC with hydrogen recycle concept (i.e. ZEG 4) with hydrogen production, CH_4 is reformed to hydrogen in the system and the net energy used in the SOFC system is reduced. The electrical efficiency is given by:

$$El_{eff} = \frac{\text{Electric energy output}}{LHV_{IN} - LHV_{OUT}}$$

When the system is fed with 1.9 times the amount of CH_4 , producing the same amount of electricity and in addition H_2 with a LHV of 257 kJ/mole, the electrical efficiency is given by:

$$El_{eff} = \frac{656 \text{ kJ / mole} * 100\%}{1.9 * 803 \text{ kJ / mole} - 0.9 * 4 * 257 * \text{kJ / mole}} = 100\%$$

The change in heating value may be converted to electricity with an electrical efficiency approaching 100%. Ohmic losses in the SOFC stacks is converted to heat and used to upgrade CH_4 to $4 * \text{H}_2$, but parasitic power and thermal losses of the system will of course reduce this efficiency. The process concept is unique with respect to efficiency, and it should also be noted that CO_2 capture and hydrogen production are inherent in the process.

In the system concept ZEG 4, NG enters the reforming&carbonation section where it is converted to H_2 , and the CO_2 is absorbed as the CaO is converted to CaCO_3 . A fraction of the H_2 goes to the H_2 cooling and compression section, while the remaining is fed into the hydrogen loop. In the hydrogen loop, the H_2 is heated before entering the SOFC stacks where only a fraction of the H_2 is used and converted to H_2O . The H_2O in $\text{H}_2/\text{H}_2\text{O}$ mixture from the SOFC stacks outlet is condensed as it is cooled down, and the H_2 is fed back to the loop. The CO_2 is released in the calcination section and enters the CO_2 cooling and compression section.

The conceptual layout of the system is shown in figure 4-7, while the overall flow diagram is shown in figure 4-8. Temperatures, flows, compositions and heat balances are given in Appendix A. The system simulation is performed for a case without H_2 recycle, hence the computed electrical efficiency may be increased further by implementation of a H_2 recycle loop.

The concept system ZEG 4 also offers great flexibility in operational conditions, compared to conventional power plants and also conventional SOFC systems, as the electrical efficiency may be increased even further at part load. The systems allow for great flexibility with respect to the produced electricity to hydrogen ratio as the production of the one or the other can be increased or decreased correspondingly to market requirements without significant effect on the overall plant efficiency.

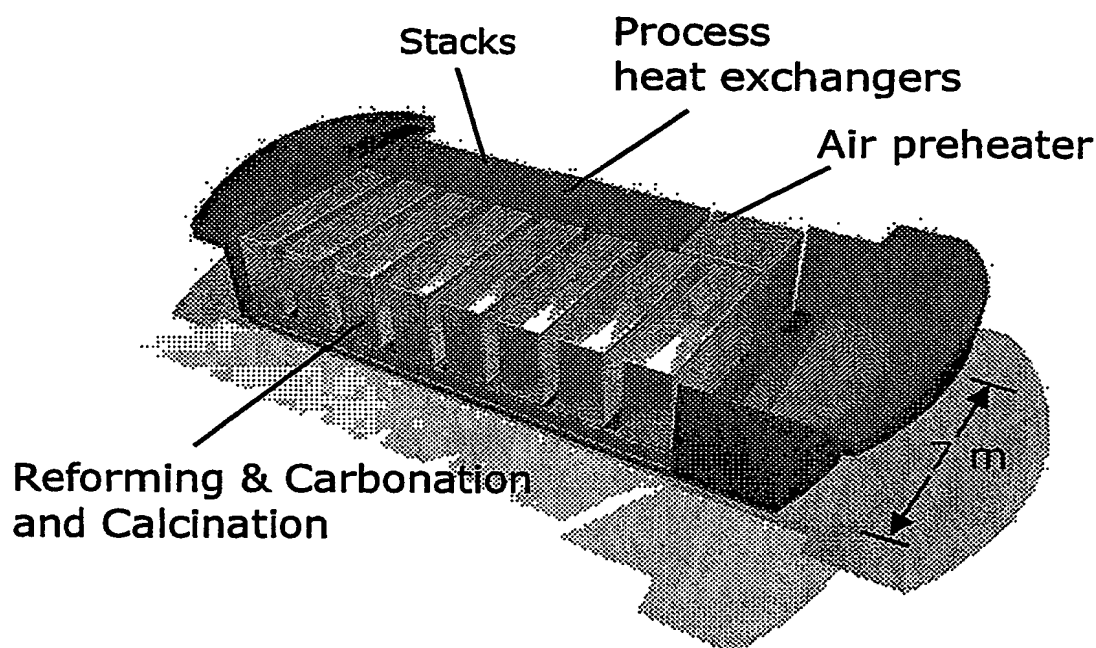


Figure 4-7 Conceptual layout of concept ZEG 4 - SOFC with Hydrogen Recycle. The shown dimensions refer to a plant with an output of 7.5 [MW_{el}] (gross DC) and 7.5 [MJ/s] of hydrogen

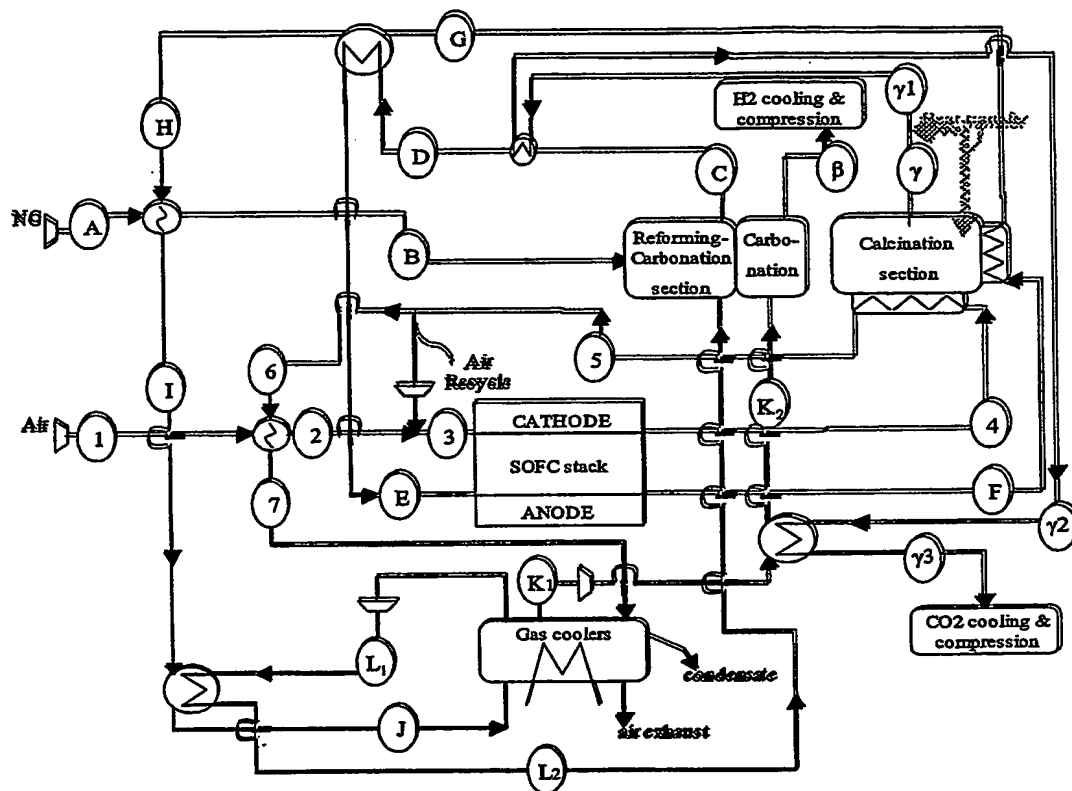



Figure 4-8 Flow diagram of ZEG 4 - SOFC with Hydrogen Recycle
 For process details see Appendix A

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| prototech  | FUTURE ENERGY PLANTS Technical Summary Report M1 5 - PROCESS SIMULATIONS | Doc.no. RP_21205_PR_01 Issue: A Date: 09.09.2002 Page: 26 of 73 |
| Project: ZEG | | |

5 PROCESS SIMULATIONS

Plant characteristics, efficiencies and power output have been estimated by process simulations evaluating heat & mass balances of the SOFC-based power plants (Appendix A). Table 5-1 shows the most relevant assumptions. Appendix A shows the operating conditions (steady state) at the most relevant points of the plants.

| Assumptions & modelling | | | | | |
|---|--------------------------------------|--------|--------------------|---------------------------------|-------------------|
| | General (if not specified in) | | | | |
| | *Particular*) ZEG 1..4 | ZEG 1 | ZEG 2 | ZEG 3 | ZEG 4 |
| Electrical output [gross DC] | 7500 kW | | | | |
| H ₂ output [LHV] | 7500 kW | | | | |
| Cell Voltage | | 0.7 V | 0.7 V | 0.7 V | 0.85 V |
| Fuel Utilization [per pass in fuel cell] | | 85 % | 85 % | 85 % | 59.60 % |
| ΔT SOFC _{stack} | | 100 °C | 100 °C | 100 °C | 110 °C |
| Prereformed methane | | 25 % | 100 % ^Δ | 100 % ^Δ | 95 % ^Δ |
| H ₂ membrane reactor: reformed methane | | - | - | 100 % ^Δ | - |
| Reforming-carbonation*: methane molar conversion | | 100 % | 100 % ^Δ | - | 95 % ^Δ |
| CO ₂ absorption rate (mol/s) | | 1.0 | 1.0 | - | 1.0 |
| CO ₂ desorption rate (mol/s) | | 1.0 | 1.0 | - | 1.0 |
| CaO conversion to CaCO ₃ | | 100 % | 100 % | - | 100 % |
| CaCO ₃ conversion to CaO | | 100 % | 100 % | - | 100 % |
| Minimum calcination temperature | | 900 °C | 900 °C | - | 850 °C |
| CaO/CO ₂ ratio (molar) | | 1.0 | 1.0 | - | 1.0 |
| H ₂ O/CH ₄ ratio (molar) at Inlet | 2 | | | | |
| Operating pressure | 1 bar | | | | |
| Pressure losses air side | 10 mbar per component; 20 mbar STACK | | | | |
| Pressure losses fuel side** | 10 mbar per component; 20 mbar STACK | | | ΔP 3bar H ₂ membrane | |

Table 5-1 Assumptions and modelling

*The reforming&carbonation reaction is $\text{CH}_4 + 2\text{H}_2\text{O} + \text{CaO} = 4\text{H}_2 + \text{CaCO}_3$

**neglected. The Natural Gas is supposed to have an outlet pressure from the grid 24bar²

In the modelling (Appendix A) the NG has been assumed to consist of 100% CH₄. Besides, if not totally prereformed ($\text{CH}_4 + \text{H}_2\text{O} \rightleftharpoons 3\text{H}_2 + \text{CO}$) the CH₄ is considered to undergo total reforming at the inlet of the Solid Oxide Fuel Cell (realistic approximation due to the high operating temperature of the SOFC stacks (950°C-1050°C)).

² according to currently operated international standard

6 EFFICIENCIES AND POWER OUTPUT

Table 6-1 reports the most relevant performance indicators. It is important to underline that the efficiency definition is based on the NET_Power_Input (LHV based) of the system. NET_Power_Input = TOT_Power_Input - H₂_Output where:

- TOT_Power_Input is the total power input LHV based [MW_{th}] of the fuel entering the system
- H₂_Output is the power output LHV based [MW_{th}] of the H₂ produced
- Net_electric_output is the gross electric output of the system minus the electric power required to drive the oxygen pump and the air, CO₂, H₂ compressors³.

The formula definitions are hereby:

$$El_{eff} = \frac{Net_electric_output}{NET_Power_Input} \quad (6.1)$$

$$Thermal_output = \frac{Hot_Water_Thermal_Output}{NET_Power_Input} \quad (6.2)$$


$$Auxiliaries = \frac{Air_compression + Power_required_by_the_oxygen_pump}{NET_Power_Input} \quad (6.3)$$

$$CO_2_compressor = \frac{CO_2_compression}{NET_Power_Input} \quad (6.4)$$

$$H_2_compressor = \frac{H_2_compression}{NET_Power_Input} \quad (6.5)$$

In the modelling the systems perform so that the gross electrical power production of the system is equal to the H₂_Output (power output LHV based [MW_{th}] of the H₂ leaving the system).

³ DC/AC conversion is not computed

| | | |
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|  | FUTURE ENERGY PLANTS Technical Summary Report M1 6 - EFFICIENCIES AND POWER OUTPUT | Doc.no. RP_21205_PR_01 |
| Project: ZEG | | Issue: A Date: 09.09.2002 Page: 28 of 73 |

| Plant configuration | ZEG 1 | ZEG 2 | ZEG 3 | ZEG 4 |
|--|-------|-------|-------|-------|
| Air mass flow @ SOFC inlet [kg/s] | 31.3 | 51.0 | 50.0 | 25.9 |
| Air recycle ratio | 2.48 | 4.68 | 4.56 | 1.45 |
| SOFC power output [MW _{el}] | 7.50 | 7.50 | 7.50 | 7.50 |
| Power required by the oxygen pump [MW _{el}] | 0.19 | 0.00 | 0.19 | 0.00 |
| Air compression [MW _{el}] | 0.29 | 0.64 | 0.48 | 0.29 |
| CO ₂ compression (55 bar) [MW _{el}] | 0.34 | 0.34 | 0.34 | 0.27 |
| H ₂ compression (350 bar) [MW _{el}] | 0.61 | 0.61 | 0.61 | 0.61 |
| Net electric output [MW _{el}] | 6.07 | 5.91 | 5.88 | 6.33 |
| H ₂ output (LHV based) [MW _{th}] | 7.50 | 7.50 | 7.50 | 7.50 |
| TOT power input (LHV) [MW _{th}] | 19.32 | 19.32 | 19.32 | 15.39 |
| NET power input (LHV) [MW _{th}] | 11.82 | 11.82 | 11.82 | 7.89 |
| LHV _{CH4} / LHV _{H2} (mol _{CH4} basis) | 0.83 | 0.83 | 0.83 | 0.83 |
| Net electrical efficiency | 0.51 | 0.50 | 0.50 | 0.80 |
| Heat to CO ₂ separation process (calcination) [MW _{th}] | 1.30 | 4.04 | 4.41* | 3.20 |
| Hot Water power output [MW _{th}] | 4.18 | 4.18 | 4.18 | 0.25 |
| Removed CO ₂ [kg/s] ** | 1.06 | 1.06 | 1.06 | 0.84 |
| CO ₂ removal efficiency [%] | ~100 | ~100 | ~100 | ~100 |

Table 6-1

Overall performances of the plants considered in the paper

*Heat to 100% reforming+shift process at 450°C

**The removed CO₂ associated to the electricity production is 0.65 [kg/s] for ZEG 1 to 3 and 0.43 [kg/s] for ZEG 4

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Figure 6-1 shows the results of the efficiencies calculations. The results can be directly computed from table 6-1 (values related to eq.(6.3) to (6.5) and to the thermal losses are showed as negative).

Referring to figure 6-1, *El eff (net)* includes power consumption for CO₂ and H₂ compression. By not compressing the H₂, for example, the *El eff* would increase 5.2% for ZEG 1, 2, 3 and 7.7% for ZEG 4.

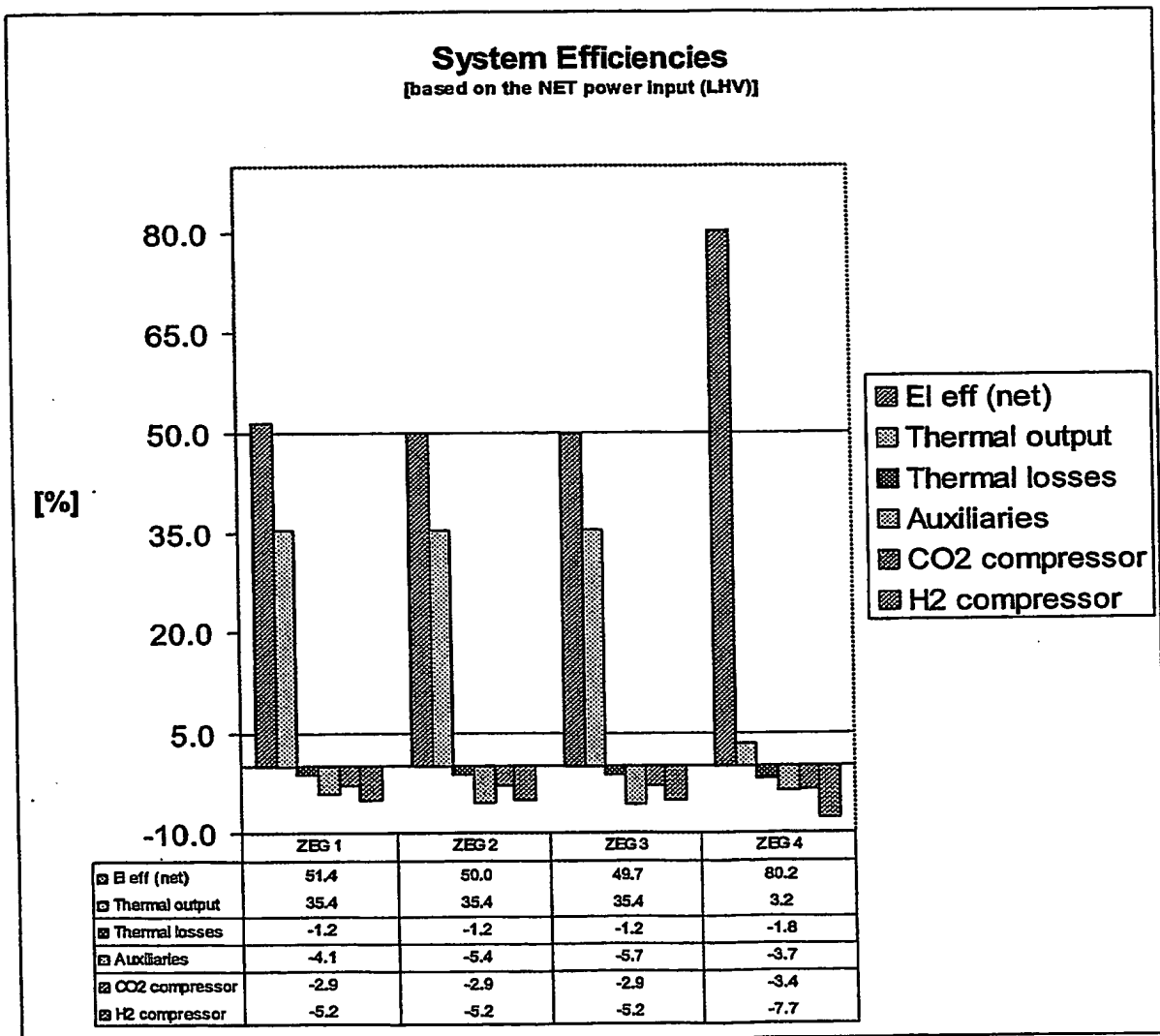



Figure 6-1 Efficiency comparison

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|  | FUTURE ENERGY PLANTS Technical Summary Report M1 | Doc.no. RP_21205_PR_01 Issue: A Date: 09.09.2002 |
| Project: ZEG | 7 - COST OF ELECTRICITY | Page: 30 of 73 |

7 COST OF ELECTRICITY

7.1 Investment costs

The investment costs analysis has been carried out following the assumptions illustrated in Table 7-1.

Costs for SOFC stacks, prereformer and CO₂ compressors have been derived from the literature [R2, R3]. H₂ compressors have been equated to CO₂ compressors in first approximation.

The cost of an oxygen pump based on a SOFC cathode-cathode technology has been calculated to be equal to a SOFC stacks with equivalent electrical power output.

The cost of a single heat exchanger has been derived calculating the amount of steel needed to exchange the thermal power. 20% of the cost found in this way has been added to include extra costs (manifolds etc.). The thermal powers are taken from the calculations (Appendix A). The minimum allowed ΔT between the average temperatures of the cold and hot sides is 50°C.

The cost of an afterburner has been estimated to be 1/10 of a power-equivalent high-temperature heat exchanger.

The cost of a H₂ membrane reformer has been considered equal to the sum of the cost of a prereformer plus the material cost (20% increased) of the palladium required (Appendix D) to filtering the H₂ from the NG-steam reforming process.

The integrated reforming&carbonation and calcination reactors have been treated as heat exchangers as a first approximation.

The scale factor has been considered to be 1 for modular subsystem, in the other cases it has been derived from the literature [R3].

The results are shown in table 7-2 and figures 7-1 and 7-2.

| Component | Scale parameter | Base specific cost, c_0 | Base size S_0 | Scale factor, f |
|--|---------------------------|---------------------------|-----------------|-------------------|
| SOFC stack | SOFC power output $_{el}$ | 150 \$/kW $_{el}$ | 250 kW $_{el}$ | 1 |
| Oxygen "pump" | SOFC power output $_{el}$ | 150 \$/kW $_{el}$ | 250 kW $_{el}$ | 1 |
| Prereformer & SOFC STACK | SOFC power output $_{el}$ | 257 \$/kW $_{el}$ | 250 kW $_{el}$ | 1 |
| H ₂ -membrane reformer & SOFC STACK | SOFC power output $_{el}$ | 265 \$/kW $_{el}$ | 250 kW $_{el}$ | 1 |
| High temperature heat exchangers (> 900°C) | Kg steel | 26 \$/kg $_{steel}$ | - | - |
| Low temperature heat exchangers | Kg steel | 13 \$/kg $_{steel}$ | - | - |
| Afterburner | Kg steel | 26 \$/kg $_{steel}$ | - | - |
| Calcination and reforming&carbonation reactors (> 900°C) | Kg steel | 26 \$/kg $_{steel}$ | - | - |
| Calcination and reforming&carbonation reactors (< 900°C) | Kg steel | 13 \$/kg $_{steel}$ | - | - |
| Compressors | Power consumed | 400 \$/kW | 30 MW | 2/3 |

N.B.:

$$C = c_0 \cdot S_0^f \cdot (S/S_0)^f$$

$f=1$ for modular subsystems

Table 7-1 Parameters adopted for investment costs

| Plant configuration | ZEG 1 | ZEG 2 | ZEG 3 | ZEG 4 |
|---|----------------------------|--------------|-------------|-------------|
| SOFC stack, M\$ | 1.13 | 1.13 | 1.13 | 1.13 |
| Prereformer, M\$ | 0.80 | - | - | - |
| H ₂ -membrane reformer, M\$ | - | - | 0.86 | - |
| Oxygen "pump", M\$ | 0.03 | - | 0.03 | - |
| Heat exchangers, M\$ | 1.61 | 1.32 | 1.20 | 1.34 |
| Afterburner, M\$ | 0.01 | 0.01 | 0.01 | - |
| CO ₂ compression, M\$ | 0.61 | 0.61 | 0.61 | 0.52 |
| H ₂ compression, M\$ | 0.89 | 0.89 | 0.89 | 0.89 |
| Other compression, M\$ | 0.54 | 0.92 | 0.76 | 0.54 |
| Calcination and reforming&carbonation reactors, M\$ | 0.16 | 1.60 | - | 1.22 |
| Process facilities | 5.78 | 6.48 | 5.49 | 5.64 |
| Contingencies | 20 % of process facilities | | | |
| Total cost [M\$] | 6.94 | 7.78 | 6.59 | 6.77 |
| El-investment costs [M\$] | 5.39 | 5.77 | 5.23 | 5.40 |
| H₂-investment costs [M\$] | 1.55 | 2.00 | 1.36 | 1.37 |
| Specific El costs [\$/kW$_{el}$] | 791 | 868 | 789 | 763 |
| Specific H₂-production costs [\$/GJ$_{H_2}$] | 8.21 | 10.57 | 7.19 | 7.23 |

Table 7-2 Estimated investment costs; better thermal integration (higher ΔT_s) and lower recycle ratios allow lower investment costs; at "Process facilities" has been added "Contingencies" according to [R4]

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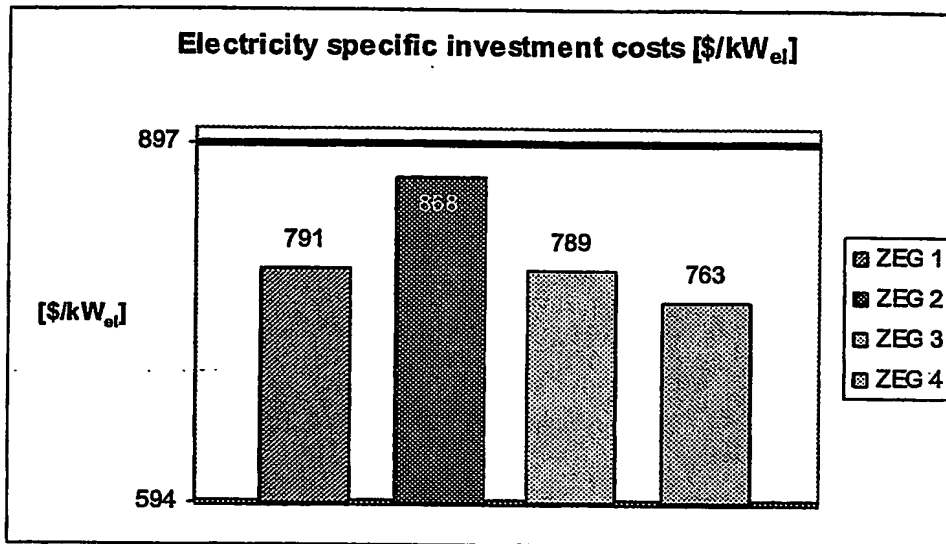


Figure 7-1 Estimated electric specific investment costs; the marked line (897 [$\$/kW_{el}$]) indicates the typical value for NGCC ("H") + flue gas CO_2 scrubber [R4]

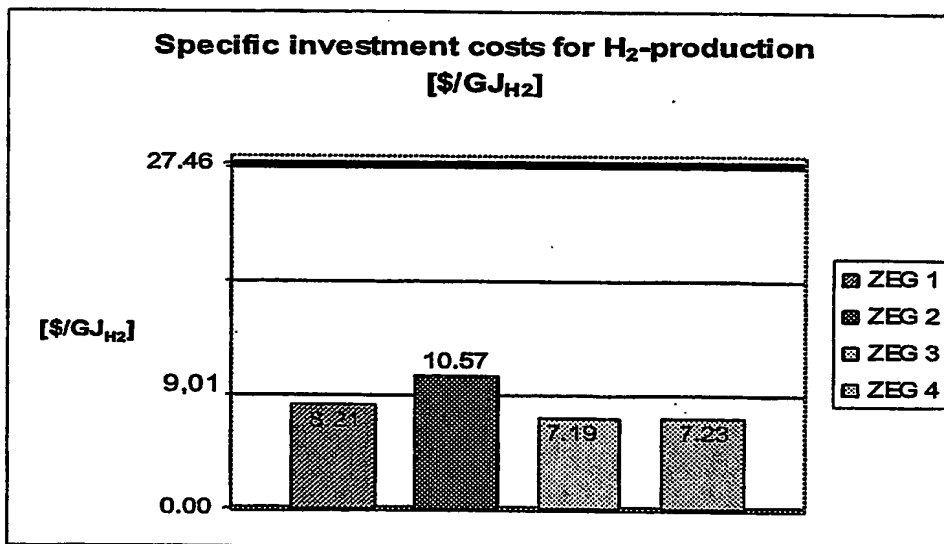


Figure 7-2 Estimated specific investment cost for H_2 -production; 9.01 [$\$/GJ_{H_2}$] indicates the typical lowest limit for Large Size Facilities Steam Methane Reforming (SMR) technology [R5]; 27.46 [$\$/GJ_{H_2}$] indicates the typical value for Small Size Facilities (i.e. ≤ 0.27 million [Nm^3/day]) Steam Methane Reforming (SMR) technology [R5]

Each system has been engineered so that the electricity gross power output equates the H₂ LHV thermal based power production. In this way it is easy to compare the investment and production costs for the systems.

Investment costs for H₂ production have been identified in:

ZEG 1:

- Reforming&carbonation and calcination reactors
- H₂ compressor
- CO₂ compressor cost directly proportional to the power required to compress the CO₂ derived from the fuel related to the production of the H₂

ZEG 2:

- part proportional to the reforming&carbonation and calcination reactors producing the H₂ destined to the system output
- H₂ compressor
- CO₂ compressor cost directly proportional to the power required to compress the CO₂ derived from the fuel related to the production of the H₂

ZEG 3:

- H₂ compressor
- CO₂ compressor cost directly proportional to the power required to compress the CO₂ derived from the fuel related to the production of the H₂

ZEG 4:

- H₂ compressor
- CO₂ compressor cost directly proportional to the power required to compress the CO₂ derived from the fuel related to the production of the H₂

In the ZEG 3 case the H₂ membrane reformer is assumed similar to a prereformer for the SOFC stacks.

7.2 Cost of Electricity (COE)

Based on the electricity generation related investment costs (figure 7-1), a preliminary estimation for the cost of electricity (COE) has been done. In Appendix B it is possible to review the economical theory that is at the basis of the calculations. The main assumptions are the following:

- 15 years plant amortization at 15% interest rate at constant currency (currency USD [\$])
- the load capacity factor of the power plant is set to 80 %
- the fuel cost (NG) is set to 3.3 \$/GJ constant over time
- due to low operating-experience for plants like the ones discussed in this paper, the O&M costs have been set to be 4% per year of investment cost

The results can be seen in figure 7-3 and show a potential for very cost efficient production of electricity and hydrogen with integrated CO₂ capture.

The higher the electrical efficiency of a system is, the lower is the sensitivity to increased fuel costs on the COE, and concept ZEG 4 may be the best solution also in respect to fuel cost sensitivity (see Appendix B, table B-4).

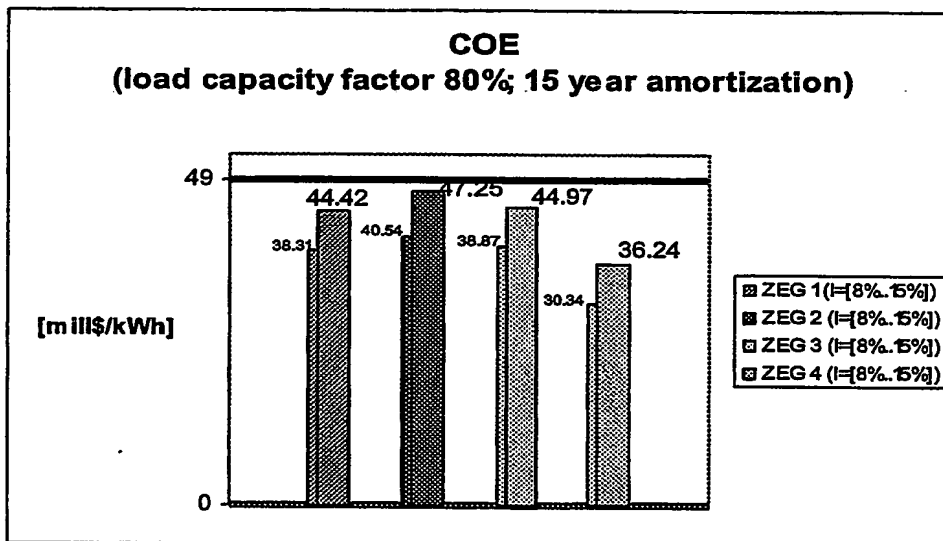


Figure 7-3 Estimated COE; for each of the ZEG concepts the interval of COE variation is showed according to the interest varying in the interval [8%..15%]; 49 [mill\$/kWh] indicates the typical value for NGCC ("H") + flue gas CO₂ scrubber (I=15%) [R4]

7.3 Cost of Hydrogen (COH₂)

Based on the specific investment costs for H₂-production (figure 7-2), a preliminary estimation for the cost of Hydrogen (COH₂) has been done (see Appendix B for more details). The main assumptions are the following:

- 15 years plant amortization at 15% interest rate at constant currency (currency USD [\$])
- the load capacity factor of the power plant is set to 80 %
- the fuel cost (NG) is set to 3.3 \$/GJ constant over time
- due to low operating-experience for plants like the ones discussed in this paper, the O&M costs have been set to be 4% per year of investment cost

The results can be seen in figure 7-4 and show that hydrogen production with integrated CO₂ capture may be performed more cost efficiently in the ZEG concepts than by conventional Steam Methane Reforming without CO₂ capture (the evaluation of the ZEG concepts is based on a Small Size Facility outline).

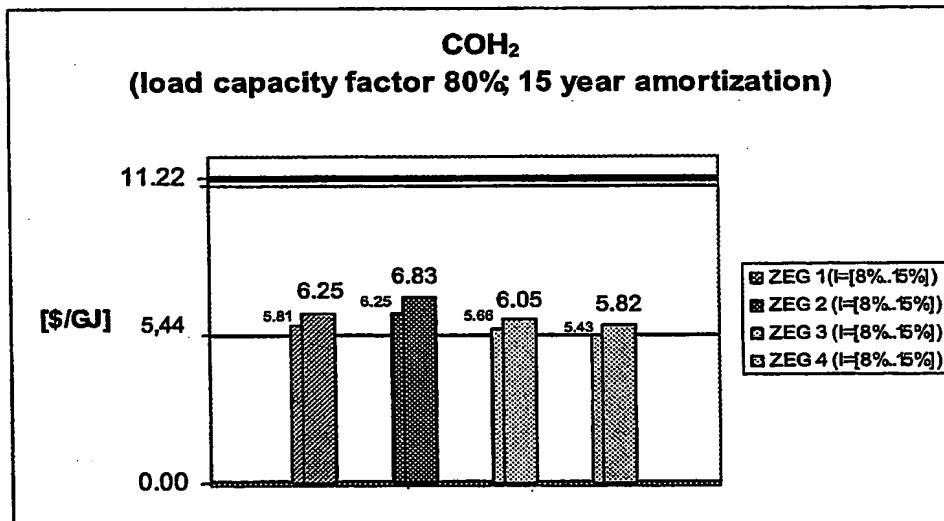



Figure 7-4 Estimated COH₂; for each of the ZEG concepts the interval of COH₂ variation is showed according to the interest varying in the interval [8%..15%]; 5.44 [\$ /GJ] indicates the typical lowest limit for Large Size Facilities Steam Methane Reforming (SMR) technology [R5]; 11.22 [\$ /GJ] indicates the typical value for Small Size Facilities (i.e. ≤0.27 million [Nm³/day]) Steam Methane Reforming (SMR) technology [R5]

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|  | FUTURE ENERGY PLANTS Technical Summary Report M1 | Doc.no. RP_21205_PR_01 |
| Project: ZEG | 7 - COST OF ELECTRICITY | Issue: A Date: 09.09.2002 |
| | | Page: 36 of 73 |

7.4 Discussion

The presented cost analysis is partly based on modular subsystems (linear scalability) and partly on not modular subsystems (i.e. compressors).

According to the presented analysis a larger plant will result in lower specific investment costs and correspondingly lower COE and COH₂.

Even considering the uncertainties of the presented analysis, the pictures given by figures 7-1 to 7-4 show that even for small facilities (7.5 [MW_{el}] electric output and 7.5 [MJ/s] hydrogen output) the COE and COH₂ of the ZEG concepts may be competitive with respectively the COE of a conventional NGCC_{GT} power plant with CO₂ scrubbing (see figure 7.3) and the COH₂ of a conventional large size facilities Steam Methane Reforming (SMR) technologies (see figure 7.4).

8 RISK FACTOR

8.1 Risk factor analysis

In order to identify the critical technologies in the described concepts, the definition of a "risk factor" (represented by a number) has been considered a good expedient. Table 8-1 shows the basic criteria for such a definition and table 8-2 shows the results obtained applying the definition itself to the components of the concepts analysed in the paper.

| <i>Risk factor legend</i> | |
|---------------------------|--------------------------------------|
| 1 | Standard tech |
| 2 | Standard tech modified |
| 3 | Under development ~ large scale demo |
| 4 | Under development ~ small scale demo |
| 5 | Dedicated tech ~ lab tested |
| 6 | Dedicated tech ~ not lab tested |

Table 8-1 Risk factor legend

| | RISK FACTOR | | |
|--|--------------------|----------------|----------------------|
| | <i>Process</i> | <i>Average</i> | <i>Physical Unit</i> |
| SOFC stack [planar technology] | 3 | 3.5 | 4 |
| Prereformer | 1 | 1 | 1 |
| H₂ membrane reformer | 2 | 3 | 4 |
| Oxygen "pump" | 3 | 3.5 | 4 |
| Heat exchangers | 1 | 1 | 1 |
| Afterburner | 1 | 1 | 1 |
| CO₂ compression | 1 | 1 | 1 |
| H₂ compression | 2 | 2 | 2 |
| Other compression | 1 | 1 | 1 |
| Calcination | 2 | 2 | 2 |
| Reforming&carbonation | 5 | 3.5 | 2 |
| System thermal integration (ZEG 1..3) | 2 | 2 | 2 |
| System thermal integration (ZEG 4) | 6 | 6 | 6 |

N.B.: the risk factor regarding the "system thermal integration" has been considered challenging for Zeg 4; it has been equated in this way to a "dedicated tech ~not lab tested"

Table 8-2 Risk factor for the components. For each of them a "Process" and a "Physical Unit" have been identified; the average value has been finally considered

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| prototech  | FUTURE ENERGY PLANTS Technical Summary Report M1 8 - RISK FACTOR | Doc.no. RP_21205_PR_01 |
| Project: ZEG | | Issue: A Date: 09.09.2002 Page: 38 of 73 |

Figure 8-1 shows the computed results for each concept. In the calculation only the components with risk factor higher than 2 have been computed because standard technology is not considered a risk parameter since the presence of a lot of "standard technology" components would reduce the final averaged risk factor. The system concepts and included technologies have been analysed with respect to technical risks, and shows that the ZEG 4 concept, identified as the most promising concept has the highest technological risk. The technical risk of this concept is mainly related to the close thermal integration required to obtain the very high efficiency; however, even with a not optimised thermal integration, this system will outperform the others.

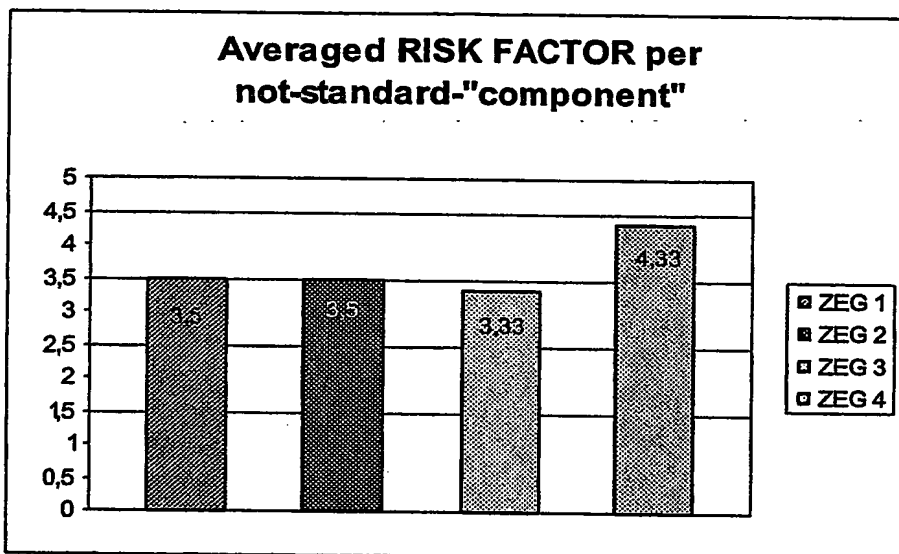


Figure 8-1 Risk factor comparison based on not-standard components

8.2 Risk factor discussion

A considerable effort is made world wide to develop and demonstrate commercial SOFC systems for power production. Two technologies are pursued for SOFC-systems; the tubular design and the planar design. The planar design allows SOFC units with higher power densities and higher efficiencies than the tubular design.


In Europe Sulzer Hexis is currently testing a number of 1 kW_e SOFC systems as a part of the planned market introduction of their unit in 2004. In the US, Siemens Westinghouse, states that market introduction of a 100 kW_e unit is planned for 2003. For the tubular design, developed by Siemens Westinghouse, significant operational has experience has been acquired in recent years.

In the present project, materials used in the planar SOFC technology are basically the same as the materials used and proven in long term testing by others.

The oxygen pump is based on an SOFC cathode-cathode planar technology and the related risk factor is thus equal to the risk factor for the SOFC.

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|  | | FUTURE ENERGY PLANTS Technical Summary Report M1 8 - RISK FACTOR | Doc.no. RP_21205_PR_01 |
| Project: ZEG | | | Issue: A Date: 09.09.2002 |
| | | | Page: 39 of 73 |

Besides this, each of the concepts discussed in the paper presents further challenges regarding the integration with a H₂ production technology including CO₂ free gas exhausts.

The integration of the reforming&carbonation reactions and reactors is one of the main challenges of the project. Based on experimental tests the two processes, implemented together, work at the investigated temperatures and pressures. The production of close to pure H₂ (95+% dry basis) with close to zero CO₂ emission (according to the reaction $\text{CH}_4 + 2\text{H}_2\text{O} + \text{CaO} = 4\text{H}_2 + \text{CaCO}_3$) has been experimentally demonstrated [R7, R8].

Further work towards optimisation is needed and an optimal reactor design has to be developed. In the next chapter possible solution are analysed.

A H₂ membrane reactor implements a classical steam-methane reforming process where the produced H₂ is filtered via a membrane. Reactors with integrated H₂-membrane are available, but up-scaling and durability still require development.

The "oxygen pump" is based on a SOFC cathode-cathode planar technology. The related risk factor is then naturally assigned.

The modality in which the high valuable heat, produced by the SOFC stacks, will be transferred to the H₂ production process is strongly dependent on the type of the reactor that will be used in the process. Under this point of view no significant problems rise in the conceptual study of ZEG 3.

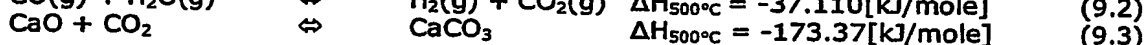
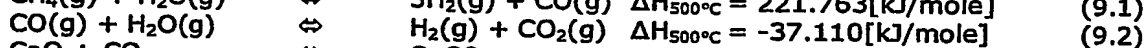
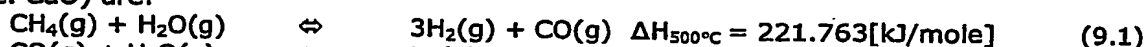
Concept ZEG 4, due to its very high efficiency, has little residual heat and the thermal system integration is a further challenge. Heat balance calculations (Appendix A) show that the thermal integration is anyway possible.

9 TECHNOLOGIES

9.1 Reforming&carbonation and calcination processes

From the risk factor analysis it is easy to see that the integrated reforming&carbonation process is one of the main challenges for concepts 1, 2 and 4.

The simultaneous reactions that occur when CH₄ is reformed in the presence of a CO₂ acceptor (i.e. CaO) are:



The combined reaction $\text{CH}_4(\text{g}) + \text{H}_2\text{O}(\text{g}) = 4\text{H}_2(\text{g}) + \text{CaCO}_3$ is almost thermally neutral (Appendix C, figure C-14).


Theoretical and experimental analysis of the two separate processes has been carried out at Prototech AS (Appendix C) in order to support the design, process simulations and cost analysis of the concepts. While Prototech has focused on the baseline reactor design (fixed bed) operated at atmospheric pressure, IFE is performing broader range of experiments to support evaluation of alternative reactor designs.

According to theoretical forecast (Appendix C, figure C-9) the carbonation process ($\text{CaO} + \text{CO}_2 \rightarrow \text{CaCO}_3$) experimentally shows (Appendix C) its potential as an intermediate step for separating the CO₂.

The reforming and carbonation processes (i.e. $\text{CH}_4 + 2\text{H}_2\text{O} + \text{CaO} = 4\text{H}_2 + \text{CaCO}_3$) implemented together gave the expected results. This has been demonstrated by IFE [R8] and by Louisiana State University [R7]. The addition of a selective CaO based acceptor to steam-methane reforming catalyst allows the reforming and shift reactions eq.(9.1) and eq.(9.2) to proceed almost to completion in a single step [R7, R8]; no shift catalyst is required. The reforming, shift, and CO₂ separation reactions are sufficiently fast that combined reaction equilibrium is closely approached over a range of pressures (ranging from 1atm [R8] to 15atm), temperatures, gas compositions, and flow rates, allowing 95+% H₂ (dry basis) to be produced in a single process vessel [R7, R8]. Only two major processing steps are needed, compared to five steps in a conventional steam-methane reforming process resulting in process simplification and improved energy efficiency [R7]. More efforts have to be done to better identify reactions rates for the integrated processes and the operational optimum in connection with the ZEG concepts.

The calcination process itself ($\text{CaCO}_3 \rightarrow \text{CaO} + \text{CO}_2$) is commercially well known. Heat has to be provided for the reaction to happen ($\Delta H_{500^\circ\text{C}} = 173.37 [\text{kJ/mole}]$). Heat balance calculations (Appendix A) show that system thermal integration is possible for all the concepts.

According to experimental results [R7] activity loss in the reforming&carbonation process can be attributed to the absorbent. According to Appendix C the CaO shows gradual loss in activity over absorption-desorption cycling of CO₂. More efforts has to be done for increasing the performance of the CaO with respect to CO₂ absorption/desorption over cycling and time. Possible need of acceptor regeneration is a step that has to be taken into account when designing the dedicated reactors. However, even in degraded state the efficiency is still high enough to be utilised in a commercial system, but improvements will reduce the size and investment costs of the reactors.

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| prototech  | FUTURE ENERGY PLANTS Technical Summary Report M1 | Doc.no. RP_21205_PR_01 |
| Project: ZEG | 9 - TECHNOLOGIES | Issue: A Date: 09.09.2002 |
| | | Page: 41 of 73 |

9.2 Reforming&carbonation and calcination reactors

In this chapter a qualitative analysis of 3 different possible reactors will be given. Different reactor concepts are shown in figures 9-1 to 9-3.

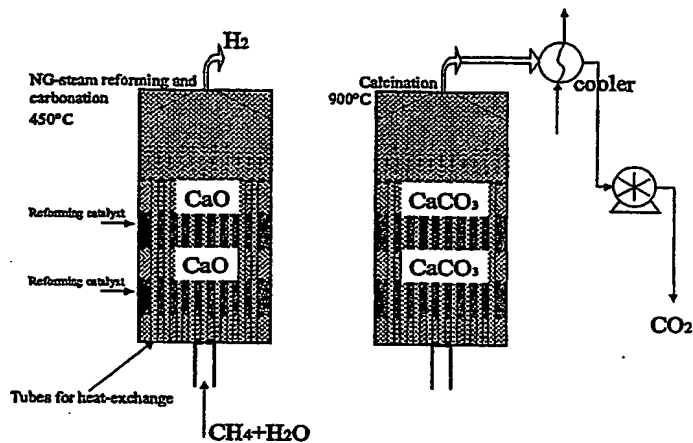


Figure 9-1 Multi-step sequential fixed bed reactors

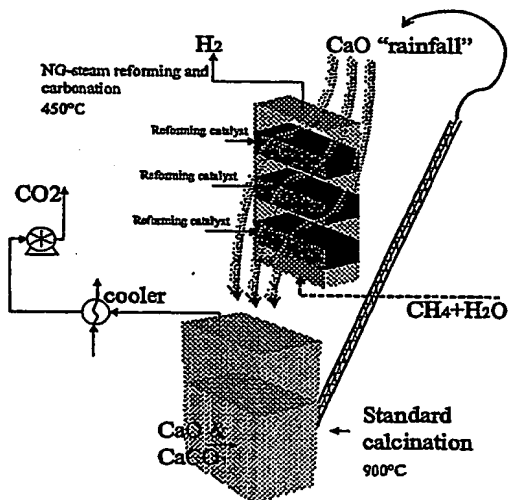


Figure 9-2 "Rain-fall" system

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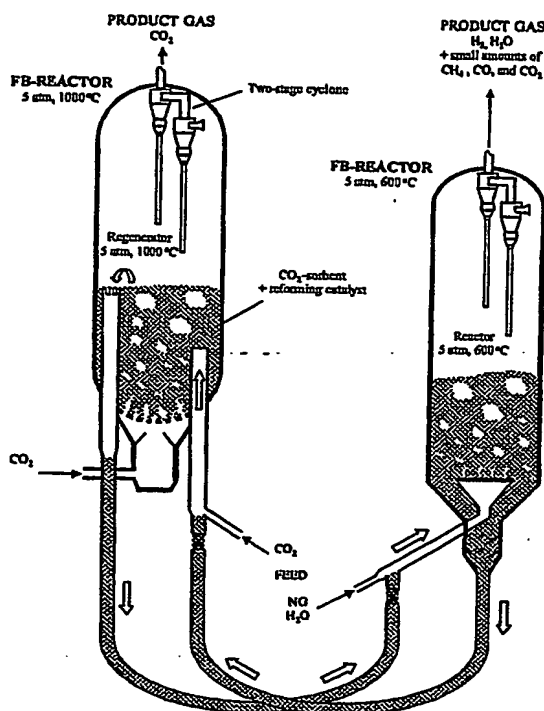


Figure 9-3 Fluidised Bed reactors [R8]


All the shown designs (figures 9-1 to 9-3) include two processes (reforming&carbonation process and calcination process) running simultaneously. As previously described the heat given to the calcination process is partly or totally recovered to drive the NG-steam reforming process via the carbonation reaction $\text{CaO} + \text{CO}_2 = \text{CaCO}_3$ (Appendix C, figure C-14). The main features of each proposal are listed below.

Multi-step sequential fixed bed reactors (figure 9-1):

- easy engineering
- contact particle-particle heat transfer
- significant radiation heat transfer in calcination reactor
- particles-fluid heat transfer
- differential pressure working possibility
- reforming catalyst and met-oxide/carbonate are kept separated (→ easy renewal if required)

"Rain fall" system (figure 9-2):

- new engineering concept for the NG-steam reforming&carbonation processes
- semi-standard engineering for the calcination reactor
- particles-fluid heat transfer
- differential pressure working possibility

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| prototech  | FUTURE ENERGY PLANTS Technical Summary Report M1 9 - TECHNOLOGIES | Doc.no. RP_21205_PR_01 |
| Project: ZEG | | Issue: A Date: 09.09.2002 |
| | | Page: 43 of 73 |

- reforming catalyst and met-oxide/carbonate are kept separated (→ easy renewal if required)

Fluidized-bed reactors (figure 9-3):

- well known advantages due to the use of fluidised-bed reactors
- reforming catalyst and met-oxide/carbonate are finely mixed; if on one side this will lead to better reaction efficiencies, on the other hand the possibility to implement a renewal step has to be investigated. The renewal may, in this case, not be necessary for the CaO/CaCO₃ (due to the use of the fluidised-bed engineering itself particles possible sintering can be avoided).

More detailed analysis on possible reactors technologies that can be considered in the optimisation study of the ZEG concepts are given by IFE [R8].

Calculations on system process integration has been carried out at Prototech AS following a batchwise project philosophy according to the multi-step sequential reactor showed in figure 9-1. The concept feasibility is supported by experimental tests [R7, R8, Appendix C]. The thermal integration feasibility is supported by the heat balance calculations reported in Appendix A.

According to experimental results and evaluating the data from [R7, R8], the pressure operation of the integrated reforming&carbonation reactor is not a critical parameter. Close to pure hydrogen can be produced (95+% dry basis) in a single step vessel over a wide range of pressures starting from 1atm and at standard⁴ reforming temperatures.

⁴ approximately 500°C

9.3 H₂ filtering

During project meetings it has been agreed that the minimum required purity of the produced H₂ should be 95% (dry basis), as a baseline. From experimental results [R7, R8] it comes to be a value directly achievable without the need of a membrane separation unit/reactor. Therefore due to the engineering of the concepts, the only case in which the produced H₂ needs a filtration step is ZEG 3. Alternative filtering possibilities has been investigated:

- palladium membrane (Pd-membrane)
- polyimide membrane
- mixed conductor membrane

Accounting for the operating temperature of the reformer catalytic-reactor (Appendix C) the best solution appears to be the use of a Pd-membrane.

From [6] there is clear evidence that a successful method for preparation of thin defect-free Pd-30%Ag membranes has been developed (membrane thickness approx. 1µm). The membranes are supported on porous ceramic substrates and show **100%** selectivity and very high hydrogen permeability in N₂+H₂ mixtures [R6]. Furthermore the use of palladium/silver alloys has overcome the problem of brittleness [R9].

In Appendix D calculations shows the theoretical area for filtering the required H₂ with a pure Pd-membrane 4µm thick operating with a ΔP of 3bar⁵. The differential pressure is an important parameter for the Pd-membrane operation (Appendix D). The higher it is the lower filtering area is required (at fixed T); similarly the lower the membrane thickness is the lower filtering area is required (at fixed T). According to [R6] the flux through the membrane will also be affected by the porous substrate on which the membrane itself will be grown. Depending on the filtering rate of the produced H₂ the retentate gas CH₄ concentration will vary; the system performance will be affected by this due to the varying extent of internal SOFC stacks reforming implying varying amounts of air requirement for cooling purposes⁶.

An optimal compromise has to be experimentally determined under operating conditions of the membrane-reformer reactor to guarantee:

- appropriate flux of H₂ through the membrane
- membrane structural-mechanical stability (which influences lifetime).

⁵ Assuming the natural gas from the grid at >24bar (according to international operated standards) no system losses for compression are involved.

⁶ In Appendix A (heat balance calculations) it has been assumed for simplicity that the H₂ production rate from the steam-methane reforming (9.1) and shift (9.2) processes and the H₂ permeation rate through the membrane are such as reactions (9.1) and (9.2) go to completion in a single step. Retentate gases and part of the H₂-filtered are fed to the SOFC stacks.


9.4 Cathode-cathode SOFC technology oxygen pump

The oxygen pump based on a SOFC cathode-cathode planar technology clearly shows the potential of being perfectly suited to our systems both physically (in terms of materials and operating conditions) and for its high efficiency. Prototech has previous experience with the manufacture and testing of oxygen pumps. Test set-ups for single cell and multi-cell testing is currently under construction.

In the figures showing the process flow schematics of the four ZEG concepts (figures 4-2, 4-4, 4-6, 4-8) the "oxygen pump" is represented (for schematic simplicity) by the presence of a N₂ stream going out from the afterburner.

Physically the oxygen pump will be integrated with the SOFC stacks.

The theoretical background for the oxygen pump calculations is shown in Appendix E. The calculations are based on experimental results from SOFC operation carried out at Prototech AS.

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|  | FUTURE ENERGY PLANTS Technical Summary Report M1 | Doc.no. RP_21205_PR_01 Issue: A Date: 09.09.2002 |
| Project: ZEG | 10 - DISCUSSION | Page: 46 of 73 |

10 DISCUSSION

10.1 Concept performances

ZEG 1 has the lowest integration between the SOFC power plant and the hydrogen production part. The thermal integration is easier than for ZEG 2 as a lower amount of solids has to be handled. This system may be operated with partial prereforming and substantial internal reforming in the SOFC stacks, hence less air recycle is needed than in ZEG 2. The performance of ZEG 1 could be raised using the exhaust heat to produce further electricity via a steam-cycle, or by utilising the excess heat to increase the hydrogen production.

In ZEG 2 the SOFC stacks is fed by H₂, resulting in greater need of air for cooling the SOFC stacks itself (no internal reforming). The higher cathode recycle ratios require higher compression works. The recycle ratio is a bit higher than for ZEG 3 due to the CO₂ capture before the SOFC stacks. The cost analyses shows that this is the most expensive solution.

ZEG 3 doesn't implement the carbonation/calcination processes and this causes a sensible reduction in investment costs. The price of a palladium-membrane, based on the material cost itself, is relatively low in terms of \$/kW_{el} (table 7-1). The development of a NG-steam reforming membrane reactor, that could in principle be fixed or fluidised, will be interesting. The implementation of an "oxygen pump" based on a SOFC cathode-cathode technology is another inspiring aspect of this concept. The membrane-reforming reactor would require pressurized fuel to operate. Assuming the grid to be at >24bar⁷ no negative implication for the system is involved. The retentate gases will have to be expanded before entering the SOFC stacks. As for ZEG 1 better performances for ZEG 2 and ZEG 3 can be achieved further using the residual heat.

ZEG 4 offers the best opportunity in terms of lower investment costs, lower COE&COH₂ and higher efficiencies. Due to its high efficiency the residual available heat is negligible, and an optimal thermal system integration is required. Recycle of H₂ is required to run the SOFC stacks close to ideal conditions and obtain a high cell voltage and appropriate fuel utilization. Due to low heat availability the carbonation process has to be totally integrated with the reforming process.

Another interesting solution would be cooling down the anode exhaust at the outlet of the SOFC stacks, absorbing the CO₂ (the process is more efficient at lower temperatures) via MgO (derived for example from an olivine mine) and depositing it as carbonate [R13]. No regeneration reactor would be needed. In this case, depending on the desired grade of purity of the produced H₂, the use of a polyimide membrane could be implemented. Cooling down the anode-exhaust to 200°C (typical working temperature of a polyimide membrane), it is possible to filter the required H₂ through the membrane and finally, after condensing the exhaust, to capture the CO₂.

All the concepts have fairly high net electrical efficiencies (NET power input LHV based, see section 6 for its definition); they are respectively of 51.4%, 50.0%, 49.7%, 80.2% (see section 6).

⁷ According to currently operated international standards

Excluding H₂ compression the computed electrical efficiencies are 56.5%, 55.2%, 54.9%, 87.9%; furthermore considering only the CO₂ compression related to the electricity production (see section 6, table 6-1) they are 57.6%, 56.3%, 56.0% and 89.6%.

The electrical efficiencies of the ZEG concepts are higher then than the one of a NGCC_{GT} with CO₂ removal that can be set (considering the "state of the art" of a "H" class GT based CC) to 52% (LHV basis) [R4].


In the case of concept ZEG 1,2 and 3, utilizing the residual heat (i.e. via steam-cycle) or implementing anode recycle [R1] the electrical efficiencies could be further increased. The residual heat could be also used for heating purposes (ZEG 1, 2 and 3).

It is possible to define the overall efficiency for one concept as the ratio between the total valuable power produced by the overall system and the total power entering the overall system⁸; the respective values are 70.2%, 69.4%, 69.3%, 89.9%⁹.

Investment costs for electricity production and for H₂ production, and the respective COE and COH₂ (figures 7-1 to 7-4), underline the market potential of the systems investigated in our study.

⁸ The computed values (from table 6-1) are LHV based and referred to the net system output. For each of the concept "valuable power produced" is meant to be the sum of the net electrical power output [MW_{el}] (see section 6, table 6-1) plus the [MJ/s] of the produced H₂; "total power entering the overall system" is meant to be the [MJ/s] of the Total fuel entering the overall system (see section 6)

⁹ For ZEG 1 to 3 they could be raised (see earlier in this heading)

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| prototech  | FUTURE ENERGY PLANTS Technical Summary Report M1 | Doc.no. RP_21205_PR_01 Issue: A Date: 09.09.2002 |
| Project: ZEG | 10 - DISCUSSION | Page: 48 of 73 |

10.2 Reforming&carbonation and calcination reactors

The baseline reactor concept is based on a gas to solid plate heat exchanger design with catalyst/absorbent in one side and heat transfer gas in the other.

Theoretical and experimental analyses, process simulations and cost analysis have shown that this baseline system is feasible, but potential improvements have been pursued, and three alternative different concepts have been discussed qualitatively in section 9.2.

Assuming that the NG from the grid is at $>24\text{bar}^{10}$, the reactors could be used pressurized with no efficiency losses for the system. In such a case an expander for the produced gas would be needed (possibility to gain power) before entering the SOFC stacks (the SOFC stacks, in our simulations, has been run at ~ 1 bar). Reactors operating pressurized will have a higher capital cost.

Anyway it has been experimentally demonstrated [R8] that a single vessel reactor running at atmospheric pressure can be used to drive the reforming&carbonation process allowing the production of close to pure hydrogen (95+% dry basis).

The calcination process itself can be run at atmospheric CO_2 pressure when the average calcination temperature is $>900^\circ\text{C}$ (see Appendix C, figure C-9).

If operating at close to atmospheric pressure the proposed way to capture the CO_2 (via met-oxide) is shown to be competitive with a conventional DEA (aqueous solution of diethanolamine) process. In our paper the capital costs for CO_2 (via met-oxide) capture vary from 0.47 to 1.51 [M\$/kg/sec] of captured CO_2 . According to a value computed from [R3] the DEA-process capital costs vary from 0.68 to 1.01 [M\$/kg/sec] of removed CO_2 .

¹⁰ According to currently operated international standard

APPENDIX A: PROCESS DETAILS

In the following tables (tables A-1 to A-4) the values of temperatures, mass flows, volume flows, compositions and thermal heat exchanged powers are shown for concept ZEG 1 to 4. The values refers directly to the *points* labelled with numbers and letters indicated in figures 4-2, 4-4, 4-6 and 4-8.

| ZEG 1 | | | | | | | | | | | | | | Loops (air&fuel) | | System(s) | |
|--------------------|------|-------|--------|----------|----|----------------|-----------------|----------------|----------------|------------------|-----------------|-------------------------------|-------------------------------|--------------------|---------|-----------|--------|
| Point | T °C | p bar | G kg/s | V m³/s | CO | O ₂ | CO ₂ | H ₂ | N ₂ | H ₂ O | CH ₄ | C ₂ H ₆ | C ₃ H ₈ | Thermal Power [MW] | error | LHV [MW] | error |
| 1 | 25 | 1 | 8.99 | 7.60 | 0 | 21 | 0 | 0 | 79 | 0 | 0 | 0 | 0 | 5.43 | | | |
| 2 | 595 | 1 | 8.99 | 22.14 | 0 | 21 | 0 | 0 | 79 | 0 | 0 | 0 | 0 | | | | |
| 3 | 850 | 1 | <31.3 | <99.73 | 0 | <21 | 0 | 0 | >79 | 0 | 0 | 0 | 0 | | | | |
| 4 | 950 | 1 | <8.99 | <31.19 | 0 | <21 | 0 | 0 | >79 | 0 | 0 | 0 | 0 | 3.67 | * | | |
| 5 | 850 | 1 | <8.99 | <28.64 | 0 | <21 | 0 | 0 | >79 | 0 | 0 | 0 | 0 | -1.05 | | | |
| 6 | 775 | 1 | <8.99 | <26.73 | 0 | <21 | 0 | 0 | >79 | 0 | 0 | 0 | 0 | -0.75 | | | |
| 7 | 670 | 1 | <8.99 | <24.05 | 0 | <21 | 0 | 0 | >79 | 0 | 0 | 0 | 0 | -1.12 | | | |
| 8 | 105 | 1 | <8.99 | <9.64 | 0 | <21 | 0 | 0 | >79 | 0 | 0 | 0 | 0 | -5.43 | | | |
| EXH | 25 | 1 | <8.99 | <7.60 | 0 | <21 | 0 | 0 | >79 | 0 | 0 | 0 | 0 | -0.73 | | 1.81 | |
| | | | | | | | | | | | | | | 0.02 | 0.22 % | | |
| A NG | 25 | 1 | 0.26 | 0.40 | 0 | 0 | 0 | 0 | 0 | 0 | 100 | 0 | 0 | | | 43.40 | |
| A FW | 25 | 1 | 0.59 | 5.88E-04 | 0 | 0 | 0 | 0 | 0 | 100 | 0 | 0 | 0 | | | | |
| B NG | 450 | 1 | 0.26 | 0.97 | 0 | 0 | 0 | 0 | 0 | 0 | 100 | 0 | 0 | 0.33 | | | |
| B FW | 450 | 1 | 0.59 | 1.94 | 0 | 0 | 0 | 0 | 0 | 100 | 0 | 0 | 0 | 2.03 | | | |
| C | 450 | 1 | 0.85 | 3.40 | 0 | 0 | 7.14 | 28.57 | 0 | 42.86 | 21.43 | 0 | 0 | | | | |
| D | 850 | 1 | 0.85 | 5.28 | 0 | 0 | 7.14 | 28.57 | 0 | 42.86 | 21.43 | 0 | 0 | 1.01 | | | |
| E | 950 | 1 | 1.75 | 8.84 | 0 | 0 | 20 | 12 | 0 | 68 | 0 | 0 | 0 | 0.36 | | 7.50 | |
| F | 720 | 1 | 1.89 | 6.65 | 0 | 0 | 20 | 0 | 0 | 80 | 0 | 0 | 0 | -0.04 | ** | 7.72 | |
| G | 200 | 1 | 1.89 | 3.17 | 0 | 0 | 20 | 0 | 0 | 80 | 0 | 0 | 0 | -1.72 | | | |
| a | 25 | 1 | 0.67 | 0.57 | 0 | 21 | 0 | 0 | 79 | 0 | 0 | 0 | 0 | | | | |
| a1 | 790 | 1 | 0.53 | 1.66 | 0 | 0 | 0 | 0 | 100 | 0 | 0 | 0 | 0 | 0.54 | | | |
| a1 | 25 | 1 | 0.53 | 0.46 | 0 | 0 | 0 | 0 | 100 | 0 | 0 | 0 | 0 | -0.47 | | | |
| EXH/y ₁ | | 1 | 1.89 | N/A | 0 | 0 | 20 | 0 | 0 | 80 | 0 | 0 | 0 | -1.97 | *** | 1.97 | |
| | | | | | | | | | | | | | | 0.07 | 1.67 % | 0.10 | 0.78 % |
| A1 NG | 25 | 1 | 0.12 | 0.19 | 0 | 0 | 0 | 0 | 0 | 0 | 100 | 0 | 0 | | | 6.22 | |
| A1 FW | 25 | 1 | 0.28 | 2.80E-04 | 0 | 0 | 0 | 0 | 0 | 100 | 0 | 0 | 0 | | | | |
| B1 NG | 450 | 1 | 0.12 | 0.44 | 0 | 0 | 0 | 0 | 0 | 0 | 100 | 0 | 0 | 0.12 | | | |
| B1 FW | 450 | 1 | 0.28 | 0.92 | 0 | 0 | 0 | 0 | 0 | 100 | 0 | 0 | 0 | 0.96 | | | |
| B | 450 | 1 | 0.06 | 1.84 | 0 | 0 | 0 | 100 | 0 | 0 | 0 | 0 | 0 | 1.42 | ^ | 1.42 | |
| B | 25 | 1 | 0.06 | 0.76 | 0 | 0 | 0 | 100 | 0 | 0 | 0 | 0 | 0 | -2.34 | **** | 2.34 | |
| y | 930 | 1 | 0.34 | 0.76 | 0 | 0 | 100 | 0 | 0 | 0 | 0 | 0 | 0 | | | | |
| y | 25 | 1 | 0.34 | 0.19 | 0 | 0 | 100 | 0 | 0 | 0 | 0 | 0 | 0 | -0.18 | ***** | | |
| | | | | | | | | | | | | | | -0.02 | -0.80 % | 0.14 | 1.87 % |

NB: 7.6 MW_{th}; AIR₀/H₂_{out} = 2 stoich_{comp}; AIR₀/FUEL₀ = 1 stoich_{comp}; H₂O/CH₄_{in} = 2; calculations LHV based.

*chemical: 100% reforming-shift Hydrogen production side (1.42MW)

*NET = 6.27MW (heat FC air-side) - 75%*Q_{reforming-shift} (Q_{reforming-shift} = 2.99MW)

**NET = 2.44MW₀ - 0.76MW₀condensing - 1.72MW₀calculation

***condensing A FW (1.63MW); total condenser: 3.24MW₀calculation

****Cooler (-0.38MW); 1.88 HHV increase from NG₀ to H₂_{out}

*****Total 0.36MW; 450C→25C 0.18MW.

Heat exchanged AIR loop/AIR in 2.61MW; afterburner power 2.44MW; reforming-shift(100%) SOFC cycle required power 2.99MW; reforming-shift(100%) H₂-production side required power 1.42MW.

Table A-1 Process details for concept ZEG 1

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ZEG 2

| Point | T °C | p bar | G kg/s | V m³/s | Molar composition, % | | | | | | | | | Loops (air&fuel) | | System | |
|---------|---------|----------|-----------|-----------|----------------------|----------------|-----------------|----------------|----------------|------------------|-----------------|-------------------------------|-------------------------------|-----------------------|---------|-------------|---------|
| | | | | | CO | O ₂ | CO ₂ | H ₂ | N ₂ | H ₂ O | CH ₄ | C ₂ H ₆ | C ₃ H ₈ | Thermal Power (MW) | error | LHV (MW) | error |
| 1 | 25 | 1 | 9 | 7.61 | 0 | 21 | 0 | 0 | 79 | 0 | 0 | 0 | 0 | | | | |
| 2 | 670 | 1 | 9 | 24.08 | 0 | 21 | 0 | 0 | 79 | 0 | 0 | 0 | 0 | 6.17 | | | |
| 3 | 850 | 1 | <51 | <162.50 | 0 | <21 | 0 | 0 | >79 | 0 | 0 | 0 | 0 | | | | |
| 4 | 950 | 1 | <51 | <176.98 | 0 | <21 | 0 | 0 | >79 | 0 | 0 | 0 | 0 | 5.99 | | | |
| 5 | 890 | 1 | <42 | <138.59 | 0 | <21 | 0 | 0 | >79 | 0 | 0 | 0 | 0 | -3.72 | | | |
| 6 | 890 | 1 | <9 | <29.70 | 0 | <21 | 0 | 0 | >79 | 0 | 0 | 0 | 0 | | | | |
| 7 | 810 | 1 | <9 | <27.66 | 0 | <21 | 0 | 0 | >79 | 0 | 0 | 0 | 0 | -0.79 | | | |
| 8 | 190 | 1 | <9 | <11.82 | 0 | <21 | 0 | 0 | >79 | 0 | 0 | 0 | 0 | -6.17 | | | |
| EXH | 25 | | | | | | | | | | | | | -1.55 | | -1.55 | |
| | | | | | | | | | | | | | | -0.07 | -0.58 % | | |
| A NG | 25 | 1 | 0.39 | 0.60 | 0 | 0 | 0 | 0 | 0 | 0 | 100 | 0 | 0 | | | 19.32 | |
| A FW | 25 | 1 | 0.87 | 8.70E-04 | 0 | 0 | 0 | 0 | 0 | 0 | 100 | 0 | 0 | | | | |
| B NG | 450 | 1 | 0.39 | 1.45 | 0 | 0 | 0 | 0 | 0 | 0 | 100 | 0 | 0 | 0.49 | | | |
| B Steam | 450 | 1 | 0.87 | 2.87 | 0 | 0 | 0 | 0 | 0 | 0 | 100 | 0 | 0 | 3.00 | | | |
| C | 450 | 1 | 0.13 | 3.85 | 0 | 0 | 0 | 100 | 0 | 0 | 0 | 0 | 0 | 1.42 | * | | |
| D | 850 | 1 | 0.13 | 5.99 | 0 | 0 | 0 | 100 | 0 | 0 | 0 | 0 | 0 | 0.79 | | | |
| E | 950 | 1 | 1.02 | 6.55 | 0 | 0 | 0 | 15 | 0 | 85 | 0 | 0 | 0 | 0.29 | | -7.50 | |
| F | 890 | 1 | 2.03 | 9.10 | 0 | 2.5 | 0 | 0 | 29 | 68.5 | 0 | 0 | 0 | 0.76 | * | | |
| G | -70 | 1 | 2.03 | N/A | 0 | 2.5 | 0 | 0 | 29 | 68.5 | 0 | 0 | 0 | | | -2.78 | |
| EXH | | 1 | 2.03 | N/A | 0 | 2.5 | 0 | 0 | 29 | 68.5 | 0 | 0 | 0 | -5.18 | ** | | |
| α | 25 | 1 | 1.01 | 0.85 | 0 | 21 | 0 | 0 | 79 | 0 | 0 | 0 | 0 | | | | |
| β | 450 | 1 | 0.06 | 1.78 | 0 | 0 | 0 | 100 | 0 | 0 | 0 | 0 | 0 | | | | |
| β | 25 | 1 | 0.06 | 0.73 | 0 | 0 | 0 | 100 | 0 | 0 | 0 | 0 | 0 | -1.66 | *** | -7.50 | |
| γ | 890 | 1 | 1.06 | 2.30 | 0 | 0 | 100 | 0 | 0 | 0 | 0 | 0 | 0 | 1.05 | | | |
| γ | 25 | 1 | 1.06 | 0.59 | 0 | 0 | 100 | 0 | 0 | 0 | 0 | 0 | 0 | -1.05 | | | |
| | | | | | | | | | | | | | | -0.09 | -1.19 % | -0.01 | -0.05 % |

NB.: 7.5 MW_{et}; AIR_{in}/H₂_{out} = 2 stoich_{max}; AIR_{in}/FUEL_{in} = 1.5 stoich_{max}; H₂O/CH₄_{in} = 2; calculations LHV based.

*chemical: molH₂_{out}/molH₂_{tot}(100%ref)*4.40MW H₂ out (HV increasing); [chemical tot: 4.40MW]

*NET = 2.44MW_{th} - 1.68MW_{ex}

**condensing FW (2.39MW); total condenser: 3.24MW_{gross}

***Cooled (-0.38MW) and LHV Increase (1.28MW@25°C) from the CH₄ that is converted to H₂_{out}

Heat exchanged AIR loop/AIR in 1.87MW; afterburner power 2.44MW; calcination section power 5.40MW; reforming-shift(100%) required power 4.40MW.

Table A-2 Process details for concept ZEG 2

ZEG 3

| Point | T °C | p bar | G kg/s | V m ³ /s | Molar composition, % | | | | | | | | | Loops (air&fuel) | | System | |
|--|---------|----------|-----------|------------------------|----------------------|----------------|-----------------|----------------|----------------|------------------|-----------------|-------------------------------|-------------------------------|-----------------------|---------|-------------|--------|
| | | | | | CO | O ₂ | CO ₂ | H ₂ | N ₂ | H ₂ O | CH ₄ | C ₂ H ₆ | C ₃ H ₈ | Thermal Power (MW) | error | LHV [MW] | error |
| 1 | 25 | 1 | 9 | 7.61 | 0 | 21 | 0 | 0 | 79 | 0 | 0 | 0 | 0 | | | | |
| 2 | 373 | 1 | 9 | 16.50 | 0 | 21 | 0 | 0 | 79 | 0 | 0 | 0 | 0 | 3.24 | | | |
| 3 | 850 | 1 | <49.97 | <159.22 | 0 | <21 | 0 | 0 | >79 | 0 | 0 | 0 | 0 | | | | |
| 4 | 950 | 1 | <49.97 | <173.40 | 0 | <21 | 0 | 0 | >79 | 0 | 0 | 0 | 0 | 5.86 | | | |
| 5 | 850 | 1 | <9 | <28.68 | 0 | <21 | 0 | 0 | >79 | 0 | 0 | 0 | 0 | -1.05 | | | |
| 6 | 770 | 1 | <9 | <26.63 | 0 | <21 | 0 | 0 | >79 | 0 | 0 | 0 | 0 | -0.83 | | | |
| 7 | 450 | 1 | <9 | <18.46 | 0 | <21 | 0 | 0 | >79 | 0 | 0 | 0 | 0 | -3.24 | | | |
| 8 | 380 | 1 | <9 | <16.68 | 0 | <21 | 0 | 0 | >79 | 0 | 0 | 0 | 0 | -0.68 | | | |
| EXH | | 1 | | | | | | | | | | | | -3.31 | | -3.31 | |
| | | | | | | | | | | | | | | -0.01 | 0.11 % | | |
| ANG | 25 | 1 | 0.39 | 0.59 | 0 | 0 | 0 | 0 | 0 | 0 | 100 | 0 | 0 | | | 15.32 | |
| AFW | 25 | 1 | 0.87 | 8.70E-04 | 0 | 0 | 0 | 0 | 0 | 100 | 0 | 0 | 0 | | | | |
| B NG | 450 | 1 | 0.39 | 1.43 | 0 | 0 | 0 | 0 | 0 | 0 | 100 | 0 | 0 | 0.49 | | | |
| B Steam | 450 | 1 | 0.87 | 2.86 | 0 | 0 | 0 | 0 | 0 | 100 | 0 | 0 | 0 | 3.00 | | | |
| C | 450 | 1 | 1.19 | 5.30 | 0 | 0 | 27 | 73 | 0 | 0 | 0 | 0 | 0 | 1.42 | * | | |
| D | 550 | 1 | 1.19 | 6.03 | 0 | 0 | 27 | 73 | 0 | 0 | 0 | 0 | 0 | 0.28 | | | |
| E | 850 | 1 | 1.19 | 8.23 | 0 | 0 | 27 | 73 | 0 | 0 | 0 | 0 | 0 | 1.05 | | -7.50 | |
| F | 950 | 1 | 2.08 | 8.97 | 0 | 0 | 27 | 11 | 0 | 62 | 0 | 0 | 0 | 0.41 | | | |
| G | 455 | 1 | 2.24 | 5.34 | 0 | 0 | 27 | 0 | 0 | 73 | 0 | 0 | 0 | -1.44 | * | | |
| H | -60 | 1 | 2.24 | N/A | 0 | 0 | 27 | 0 | 0 | 73 | 0 | 0 | 0 | | | -0.77 | |
| EXH | | 1 | | | | | | | | | | | | -3.69 | ** | | |
| α | 25 | 1 | 1.01 | 0.85 | 0 | 21 | 0 | 0 | 79 | 0 | 0 | 0 | 0 | | | | |
| β | 450 | 1 | 0.06 | 1.78 | 0 | 0 | 0 | 100 | 0 | 0 | 0 | 0 | 0 | | | -7.50 | |
| β | 25 | 1 | 0.06 | 0.73 | 0 | 0 | 0 | 100 | 0 | 0 | 0 | 0 | 0 | -1.66 | *** | | |
| α | 790 | 1 | 0.53 | 1.65 | 0 | 21 | 0 | 0 | 79 | 0 | 0 | 0 | 0 | 0.54 | | | |
| α1 | 25 | 1 | 0.53 | 0.46 | 0 | 0 | 0 | 0 | 100 | 0 | 0 | 0 | 0 | -0.47 | | | |
| NB: 7.5 MWel; AIR _{ref} /H ₂ _{ref} = 2 stoich _{concept} ; AIR _{ref} /FUEL _{ref} = 1 stoich _{concept} ; H ₂ O/CH ₄ _{ref} = 2; calculations LHV based. | | | | | | | | | | | | | | -0.07 | -0.97 % | 0.24 | 1.26 % |

NB: 7.5 MWel; AIR_{ref}/H₂_{ref} = 2 stoich_{concept}; AIR_{ref}/FUEL_{ref} = 1 stoich_{concept}; H₂O/CH₄_{ref} = 2; calculations LHV based.

*chemical: molH₂ out/molH₂ in (100%ref) 4.40MW H₂ out (HV increasing); [chemical tot: 4.40MW]

*NET = 2.44MW_{el} - 3.60MW_{el} reforming - 0.28MW_{el} super-heating

**condensing FW (2.39MW); total condenser: 3.24MW_{heat}

***Cooler (-0.38MW) and LHV increase (1.28MW@25°C) from the CH₄ that is converted to H₂ out

Heat exchanged AIR loop/AIR in 4.61MW; afterburner power 2.44MW; reforming-shift(100%) required power 4.40MW.

Table A-3 Process details for concept ZEG 3

ZEG 4

| ZEG 4 | | | | | | | | | | | | | | | Loops (air & fuel) | | System | |
|---|------|-------|--------|----------|----------------------|----------------|-----------------|----------------|----------------|------------------|-----------------|-------------------------------|-------------------------------|--------------------|--------------------|----------|--------|--------|
| Point | T °C | p bar | G kg/s | V m³/s | Molar composition, % | | | | | | | | | Thermal Power (MW) | error | LHV (MW) | error | |
| | | | | | CO | O ₂ | CO ₂ | H ₂ | N ₂ | H ₂ O | CH ₄ | C ₂ H ₆ | C ₃ H ₈ | | | | | |
| 1 | 25 | 1 | 10.56 | 8.93 | 0 | 21 | 0 | 0 | 79 | 0 | 0 | 0 | 0 | | | | | |
| 2 | 850 | 1 | 10.56 | 33.65 | 0 | 21 | 0 | 0 | 79 | 0 | 0 | 0 | 0 | 9.47 | | | | |
| 3 | 850 | 1 | <25.87 | <82.43 | 0 | <21 | 0 | 0 | >79 | 0 | 0 | 0 | 0 | | | | | |
| 4 | 960 | 1 | <25.88 | <90.51 | 0 | <21 | 0 | 0 | >79 | 0 | 0 | 0 | 0 | | | | | |
| 5 | 850 | 1 | <25.89 | <82.43 | 0 | <21 | 0 | 0 | >79 | 0 | 0 | 0 | 0 | 3.34 | | | | |
| 6 | 850 | 1 | <10.56 | <33.65 | 0 | <21 | 0 | 0 | >79 | 0 | 0 | 0 | 0 | -3.34 | | | | |
| 7 | 25 | 1 | <10.56 | <8.93 | 0 | <21 | 0 | 0 | >79 | 0 | 0 | 0 | 0 | | | | | |
| EXH | | | | | | | | | | | | | | -9.47 | | | | |
| | | | | | | | | | | | | | | 0.00 | | | | |
| ANG | 25 | 1 | 0.31 | 0.47 | 0 | 0 | 0 | 0 | 0 | 0 | 100 | 0 | 0 | 0.00 | 0.00 % | | | |
| B NG | 450 | 1 | 0.31 | 1.15 | 0 | 0 | 0 | 0 | 0 | 0 | 100 | 0 | 0 | | | 15.39 | | |
| C | 450 | 1 | 0.196 | 4.49 | 0 | 0 | 0 | 96.20 | 0 | 2.53 | 1.27 | 0 | 0 | 0.39 | | | | |
| D | 550 | 1 | 0.196 | 5.11 | 0 | 0 | 0 | 96.20 | 0 | 2.53 | 1.27 | 0 | 0 | 1.43 | A | | | |
| E | 850 | 1 | 0.196 | 6.98 | 0 | 0 | 0 | 96.20 | 0 | 2.53 | 1.27 | 0 | 0 | 0.23 | | | | |
| F | 960 | 1 | 0.927 | 7.86 | 0 | 0 | 1.23 | 39.90 | 0 | 58.86 | 0 | 0 | 0 | 0.71 | | | | |
| G | 850 | 1 | 0.927 | 7.15 | 0 | 0 | 1.23 | 39.90 | 0 | 58.86 | 0 | 0 | 0 | 0.33 | | 7.5 | | |
| H | 605 | 1 | 0.927 | 5.59 | 0 | 0 | 1.23 | 39.90 | 0 | 58.86 | 0 | 0 | 0 | -0.33 | | | | |
| I | 465 | 1 | 0.927 | 4.70 | 0 | 0 | 1.23 | 39.90 | 0 | 58.86 | 0 | 0 | 0 | -0.71 | | | | |
| J | -40 | 1 | 0.927 | N/A | 0 | 0 | 1.23 | 39.90 | 0 | 58.86 | 0 | 0 | 0 | -0.39 | | | | |
| K ₁ | 25 | 1 | 0.104 | 0.78 | 0 | 0 | 3 | 97 | 0 | 0 | 0 | 0 | 0 | -2.39 | | | | |
| K ₂ | 450 | 1 | 0.104 | 1.90 | 0 | 0 | 3 | 97 | 0 | 0 | 0 | 0 | 0 | | | | | |
| L ₁ | 25 | 1 | 0.691 | 6.91E-04 | 0 | 0 | 0 | 0 | 0 | 100 | 0 | 0 | 0 | 0.40 | | | | |
| L ₂ | 450 | 1 | 0.691 | 2.27 | 0 | 0 | 0 | 0 | 0 | 100 | 0 | 0 | 0 | | | | | |
| EXH | | | 0.133 | | 0 | 0 | 0 | 0 | 0 | 100 | 0 | 0 | 0 | 2.39 | | | | |
| Y | 900 | 1 | 0.844 | 1.85 | 0 | 0 | 100 | 0 | 0 | 0 | 0 | 0 | 0 | -0.52 | * | | | |
| Y ₁ | 670 | 1 | 0.844 | 1.48 | 0 | 0 | 100 | 0 | 0 | 0 | 0 | 0 | 0 | 0.85 | | | | |
| Y ₂ | 450 | 1 | 0.844 | 1.14 | 0 | 0 | 100 | 0 | 0 | 0 | 0 | 0 | 0 | -0.23 | | | | |
| Y ₃ | 25 | 1 | 0.844 | 0.47 | 0 | 0 | 100 | 0 | 0 | 0 | 0 | 0 | 0 | -0.23 | | | | |
| β | 450 | 1 | 0.062 | 1.84 | 0 | 0 | 0 | 100 | 0 | 0 | 0 | 0 | 0 | -0.38 | | | | |
| β | 25 | 1 | 0.062 | 0.76 | 0 | 0 | 0 | 100 | 0 | 0 | 0 | 0 | 0 | | | 7.5 | | |
| | | | | | | | | | | | | | | -1.66 | ** | -0.25 | | |
| NB: 7.5 MW _{net} , AIR/H ₂ = 2 stoich, H ₂ O/CH ₄ = 2; calculations LHV based. | | | | | | | | | | | | | | | -0.11 | -1.61 % | -0.14 | 0.92 % |
| *chemical mol _{in} / mol _{out} = 3.51 MW _{net} air / GW _{th} methane; **chemical mol _{in} / mol _{out} = 3.51 MW _{net} air / GW _{th} methane | | | | | | | | | | | | | | | | | | |

NB: 7.5 MW_{tot}; AIR_{in}/H₂_{out} = 2 stoich_{top}; H₂O/CH₄_{in} = 2; calculations LHV based.

*chemical: mol_{air}/mol_{CH₄} = 3.51MW H₂ out (HV increasing); 3.51MW tot (100% reforming shift); 5%_{reforming} happens in the SOFC

*NOT condensing out water (LHV based calculations)

*Cooling (-0.38MW) and LHV increase (1.28MW@25°C) from the CH₄_{in} that is converted to H₂_{out}

Heat exchanged AIR loop/AIR in 0.00MW; PRE reforming shift(95%) required power 3.40MW.

Table A-4 Process details for concept ZEG 4

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APPENDIX B: COST ANALYSIS

The detailed economical evaluation of ZEG 4 is reported; for the other concepts the same outline has been followed.

Assuming:

Operating parameters

| O&M (cost per year of capital) | Fuel cost [\$/GJ] (constant over years) | Return on capital [%] | Taxes on income | Plant life (value 0 at the end) | Years amortization [yr] | Interest rate [%] | Load Capacity Factor | El _{eff} * |
|--------------------------------|---|-----------------------|-----------------|---------------------------------|-------------------------|-------------------|----------------------|---------------------|
| 4 % | 3.3 | 0 % | 28 % | 15 | 15 | 15 % | 80 % | 0.90 |


Table B-1 **Operating parameters**
 *see section 10.1

The *COE due to fuel* take into account the expenses related to the NET power input of the system (see section 6). The rest of the fuel power input of the system is associated with the *COH₂ due to fuel*. The *COH₂* includes the cost for compressing the produced H₂ and the cost for compressing the CO₂ associated to the H₂ production process. The calculated *total COE* is used to compute the *COH₂ due to electricity*.

Results (for ZEG 4) are reported in the following tables B-2 and B-3.

Tables B-4 and B-5 show the computed values for all the concepts (ZEG 1 to 4) assuming the following parameters (see section 7):

- 15 years plant amortization at 15% interest rate at constant currency (currency USD [\$])
- the load capacity factor of the power plant is set to 80 %
- the fuel cost (NG) is set to 3.3 \$/GJ constant over time
- due to low operating-experience for plants like the ones discussed in this paper, the O&M costs have been set to 4% per year of investment cost

| | | |
|--|---|---|
| prototech  | FUTURE ENERGY PLANTS Technical Summary Report M1 Appendix B: - COST ANALYSIS | Doc.no. RP_21205_PR_01 |
| Project: ZEG | | Issue: A Date: 09.09.2002 |
| | | Page: 54 of 73 |

ZEG 4 (EL-prod)

| | | | |
|---------------------------------|---|------------------|--------------------|
| Assume: Debt | | 5398504 \$ | (Investment costs) |
| Total Capital | | 5398504 \$ | |
| Return on total capital | | 0 \$ | |
| | 0 % of Total Capital | | |
| + Income Taxes | | | |
| | 28 % of Profit Before Taxes | 0 \$ | |
| Required Profit Before Taxes | | 0 \$ | |
| + Principal and Interest | | | |
| | 15 % interest on debt & 15 year amortization | 923236 \$ | |
| + O&M | | | |
| | 4 % of Total Capital | 215940 \$ | |
| + Fuel | | | |
| | 3.3 \$/GJ & 1 year operation | 657003 \$ | |
| = Total Annual Required Revenue | | 1796180 \$ | |
| | 1) COE due to amortization | 18.63 mill\$/kWh | |
| | 2) COE due to O&M | 4.36 mill\$/kWh | |
| | 3) COE due to fuel | 13.26 mill\$/kWh | |
| | COE (1+2+3) | 36.24 mill\$/kWh | |
| | TOT COE | 36.24 mill\$/kWh | |
| Profit Before Taxes | | 0.0 M\$ | |

Table B-2 ZEG 4 Cost of Electricity evaluation

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ZEG 4 (H₂-prod)

| | | | |
|---------------------------------|---|------------|--------------------|
| Assume: Debt | | 1368000 \$ | (investment costs) |
| Total Capital | | 1368000 \$ | |
| Return on total capital | | 0 \$ | |
| | 0 % of Total Capital | | |
| + Income Taxes | | 0 \$ | |
| | 28 % of Profit Before Taxes | | |
| Required Profit Before Taxes | | 0 \$ | |
| + Principal and Interest | | | |
| | 15 % Interest on debt & 15 year amortization | 233951 \$ | |
| + O&M | | | |
| | 4 % of Total Capital | 54720 \$ | |
| + El for compression | | | |
| | 36.24 mill\$/kWh | 188330 \$ | |
| + Fuel | | | |
| | 3.3 \$/GJ & 1 year operation | 623924 \$ | |
| = Total Annual Required Revenue | | 1100925 \$ | |
| | 1) COH ₂ due to amortization | 1.24 \$/GJ | |
| | 2) COH ₂ due to O&M | 0.29 \$/GJ | |
| | 2) COH ₂ due to El | 1.00 \$/GJ | |
| | 4) COH ₂ due to fuel | 3.30 \$/GJ | |
| | COH ₂ (1+2+3+4) | 5.82 \$/GJ | |
| | TOT COH ₂ | 5.82 \$/GJ | |
| Profit Before Taxes | | 0.0 M\$ | |

Table B-3 ZEG 4 Cost of H₂ evaluation

No returns on the capital investment have been taken into account, hence the debt equates the total capital.

Figures B-1 to B-2 and B-4 to B-5 show the varying of the COE and of the COH₂ when varying the interest rate and years amortisation.

Figures B-3 and B-6 shows the variation of the payback period as a function of the selling prices; higher selling prices → shorter payback → higher profits.

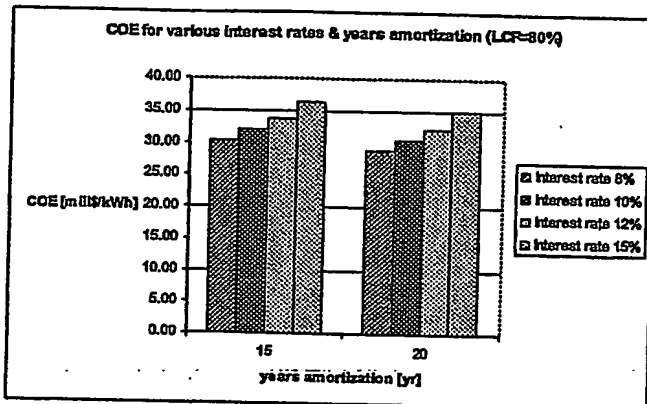


Figure B-1 COE for various interest rates and years of amortisation

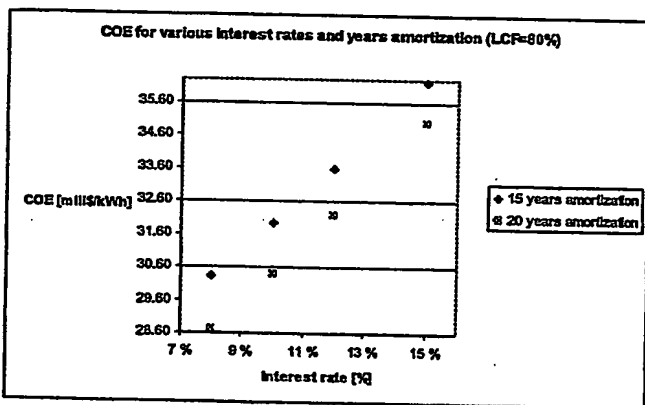


Figure B-2 COE for various interest rates and years of amortisation

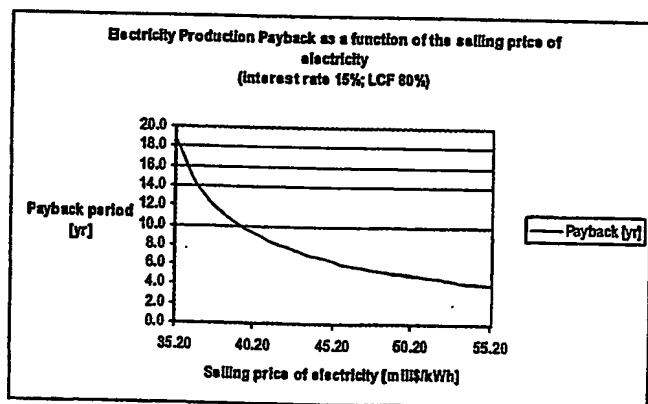


Figure B-3 Electricity production payback period as a function of the electricity selling price

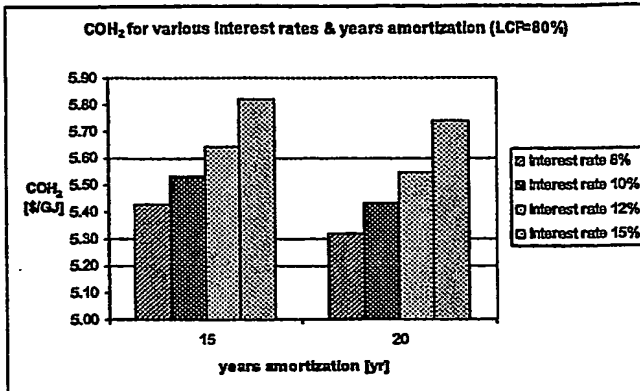


Figure B-4 CO₂ for various interest rates and years of amortisation

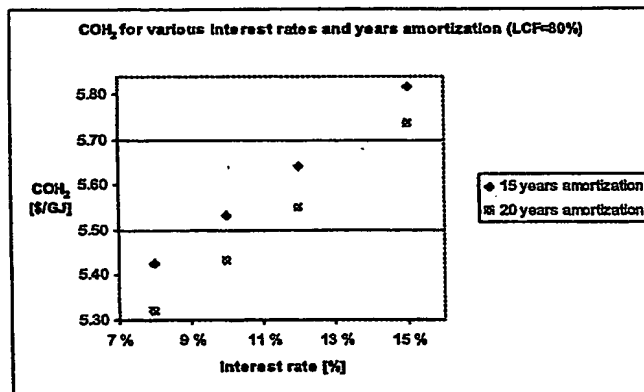


Figure B-5 CO₂ for various interest rates and years of amortisation

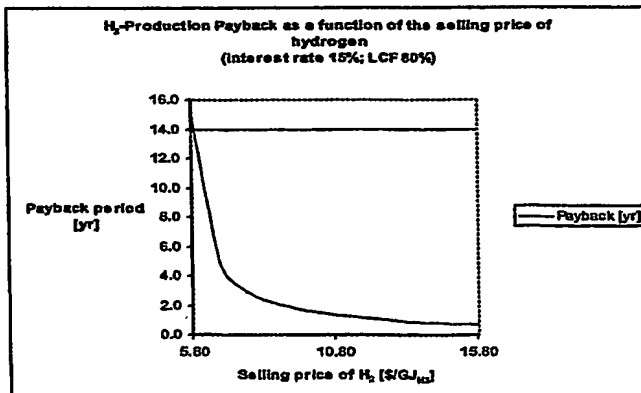



Figure B-6 H₂ production payback period as a function of the H₂ selling price

| | | |
|---|---|---|
|  | FUTURE ENERGY PLANTS Technical Summary Report M1 | Doc.no. RP_21205_PR_01 |
| Project: ZEG | Appendix B: - COST ANALYSIS | Issue: A Date: 09.09.2002 Page: 58 of 73 |

| Electricity production | | | | |
|---|-------|-------|-------|-------|
| Plant configuration | ZEG 1 | ZEG 2 | ZEG 3 | ZEG 4 |
| COE due to plant* amortization [mill\$/kWh] | 19.30 | 21.18 | 19.26 | 18.63 |
| COE due to O&M* [mill\$/kWh] | 4.51 | 4.95 | 4.51 | 4.36 |
| COE due to fuel* [mill\$/kWh] | 20.61 | 21.11 | 21.20 | 13.26 |
| Total COE [mill\$/kWh] | 44.42 | 47.25 | 44.97 | 36.24 |

*part related to the electricity production

Table B-4 **Estimated COE (Cost of Electricity) for the ZEG concepts (15 years plant amortization, 15% interest rate at constant currency (currency USD [\$]), 80% load capacity factor, 3.3 [\$/GJ] fuel cost (NG) constant over time), O&M costs set to 4% per year of investment cost).**

| Hydrogen production | | | | |
|--|-------|-------|-------|-------|
| Plant configuration | ZEG 1 | ZEG 2 | ZEG 3 | ZEG 4 |
| COH ₂ due to plant** amortization [\$/GJ] | 1.40 | 1.81 | 1.23 | 1.24 |
| COH ₂ due to O&M** [\$/GJ] | 0.33 | 0.42 | 0.29 | 0.29 |
| COH ₂ due to electricity [\$/GJ] | 1.22 | 1.30 | 1.24 | 1.00 |
| COH ₂ due to fuel** [\$/GJ] | 3.30 | 3.30 | 3.30 | 3.30 |
| Total COH ₂ [\$/GJ] | 6.25 | 6.83 | 6.05 | 5.82 |

**part related to the hydrogen production

Table B-5 **Estimated COH₂ (Cost of Hydrogen) for the ZEG concepts (15 years plant amortization, 15% interest rate at constant currency (currency USD [\$]), 80% load capacity factor, 3.3 [\$/GJ] fuel cost (NG) constant over time), O&M costs set to 4% per year of investment cost).**

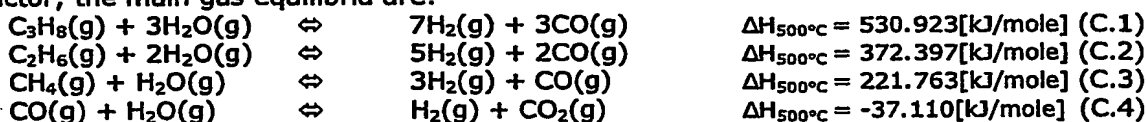
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APPENDIX C: NG-WATER REFORMING, CARBONATION AND CALCINATION

C1 NATURAL GAS-WATER REFORMING

The reforming experiments have been carried out in the presence of a catalyst. Considering the Natural Gas reforming several reactions take place when the NG goes through the catalytic reactor; the main gas equilibria are:



Figures C-1 to C-4 show the temperature dependence of the K_p of (C.1), (C.2), (C.3) and (C.4).

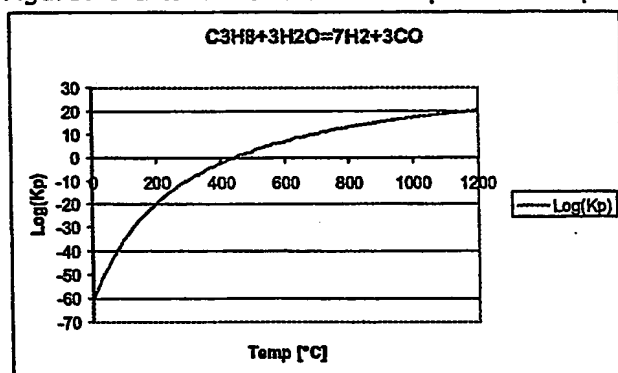


Figure C-1 $\text{Log}(K_p)$ as a function of the temperature for the reaction $C_3H_8(g) + 3H_2O(g) = 7H_2(g) + 3CO(g)$

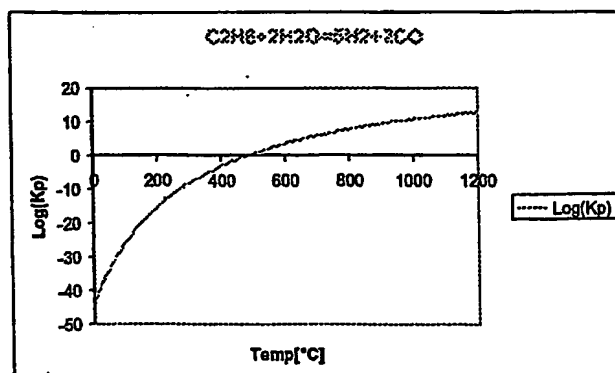


Figure C-2 $\text{Log}(K_p)$ as a function of the temperature for the reaction $C_2H_6(g) + 2H_2O(g) = 5H_2(g) + 2CO(g)$

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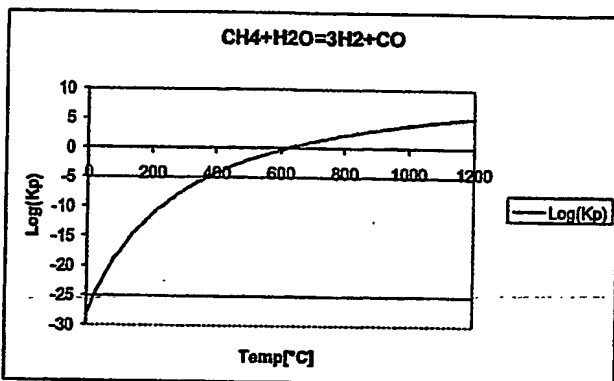


Figure C-3 **Log(K_p) as a function of the temperature for the reaction**
CH₄(g)+H₂O(g)=3H₂(g)+CO(g)

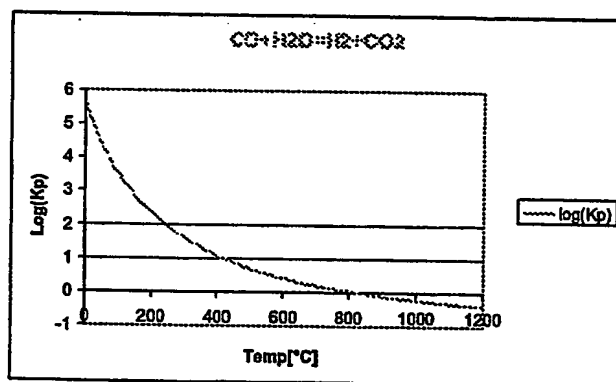
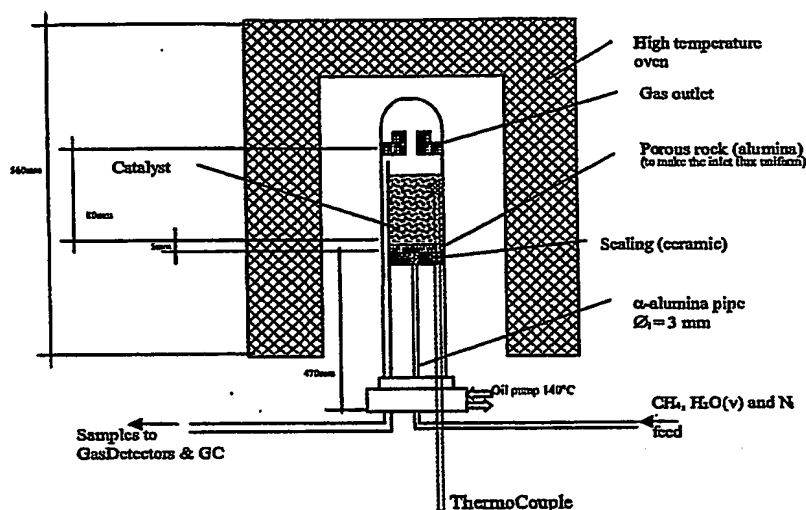


Figure C-4 **Log(K_p) as a function of the temperature for the reaction**
CO(g)+H₂O(g)=H₂(g)+CO₂(g)

C1.1 Experimental Setup

The experimental set-up is shown in figure C-5 and the explanation follows.



$ID_{reactor} = 3.4 \text{ [cm]}$

Figure C-5 Experimental set-up.

An alumina tube-continuous-reactor operating at atmospheric pressure in a high temperature-controlled oven has been used.


The catalyst type was RKNR 4,5x4,5 from Haldor Topsøe a.s.; it consisted of finely divided nickel on magnesia (MgO) support; the basic macro-shape was cylindrical with height 4,5mm and OD 4,5mm.

The feed was composed by water vapour, natural gas (or methane) and nitrogen.

The natural gas composition was:

| | |
|-------------------------------|--------------|
| N ₂ | 1.28% (vol.) |
| CO ₂ | 0.97% |
| CH ₄ | 84.3% |
| C ₂ H ₆ | 12.46% |
| C ₃ H ₈ | 0.89% |

The feed was supplied by means of mass flowmeters.

| | | |
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|  | FUTURE ENERGY PLANTS Technical Summary Report M1 Appendix C: - NG-WATER REFORMING, CARBONATION and CALCINATION | Doc.no. RP_21205_PR_01 |
| Project: ZEG | | Issue: A Date: 09.09.2002 Page: 62 of 73 |

The nitrogen has been used during the heating up period and the shut down one, according to the catalyst specification (not oxidizing atmosphere).

The water vapour was fed via a controlled steamer.

An oil pump has been used to maintain the temperature at the base of the reactor (to avoid water condensation).

The temperature inside the reactor (and the catalyst itself) has been measured by means of a K-type thermocouple.

The sampling has been done after the end of the catalyst bed; the water has been condensed before instrumental detection of the sampling.

The instrumental detection has been carried out using IR Gas Detectors (for CH₄, CO₂ and CO) and a Gas Chromatograph HP5890 SERIES II opportunely calibrated.

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C1.2 Experimental results and conclusions

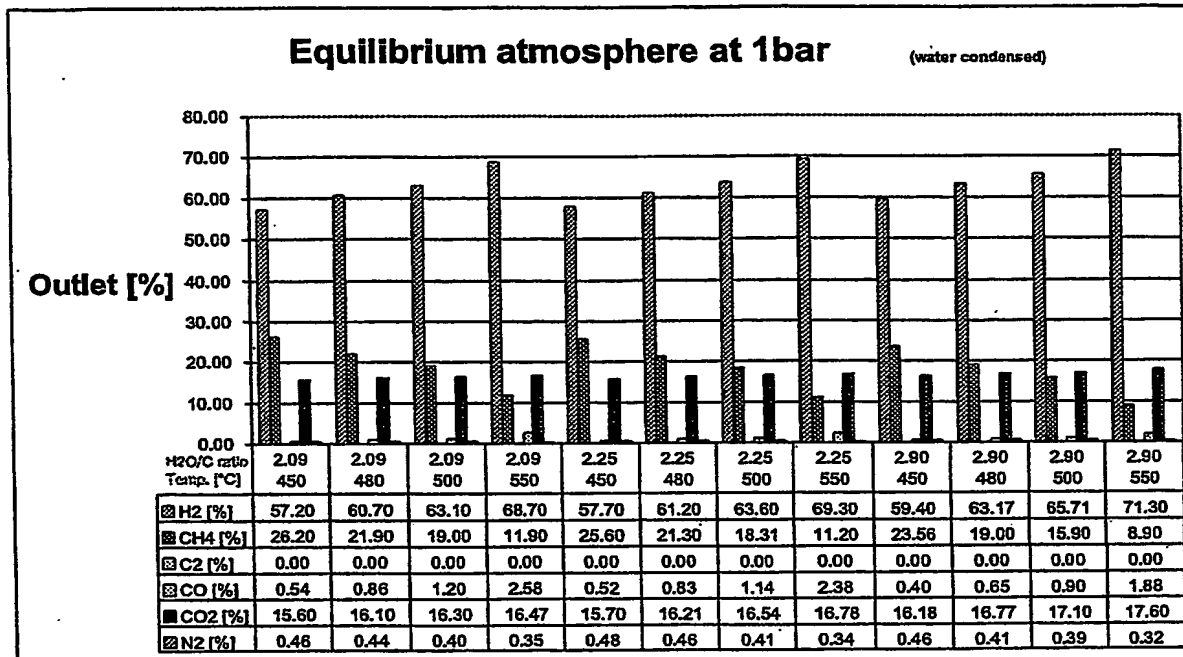


Figure C-6 Equilibrium atmosphere (dry) at 1 bar

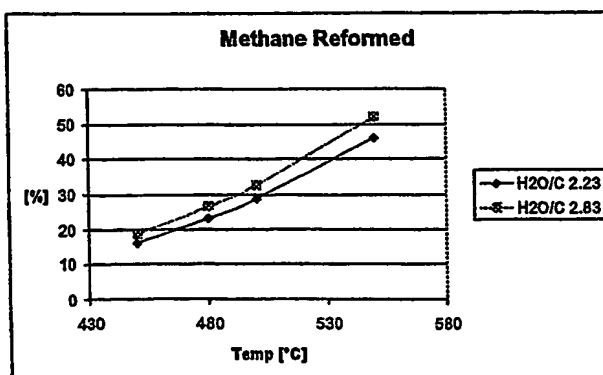



Figure C-7 % of methane reformed as a function of the temperature and the H₂O/C ratio at the inlet (P_{tot}=1 bar)

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|  | FUTURE ENERGY PLANTS Technical Summary Report M1 Appendix C: - NG-WATER REFORMING, CARBONATION and CALCINATION | Doc.no. RP_21205_PR_01 Issue: A Date: 09.09.2002 Page: 64 of 73 |
| Project: ZEG | | |

The main conclusions of the study can be summarized as follow:

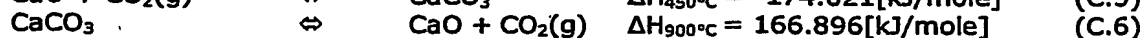
- The most significant parameters are the temperature and the steam to carbon ratio at the inlet
- Equilibrium conditions (at the investigated pressure and temperatures) are reached in a negligible time when the reactor and the catalyst have reached the set temperature
- 0.1 g catalyst per ml/min natural gas is *sufficient* (it is not a lower limit) to reform all the propane and all the ethane at temperature higher than 450°C and with a H₂O/C ratio of 2.09 or higher.
- The reforming should be operated at or above 450°C with a steam to carbon ratio at the inlet of 2.09 or higher to be sure to avoid coke deposition.
- The RKNGR 4,5x4,5 catalyst performance are not significantly affected by one shut down if the requirements given by Haldor Topsøe a.s. are followed.

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C2 CARBONATION AND CALCINATION CYCLING

The investigated carbonation and calcination cyclically performed reactions are



Figures C-8 and C-9 show the temperature dependence of the K_p of (C.5) and (C.6). The equilibrium plots are valid when all 3 components are present

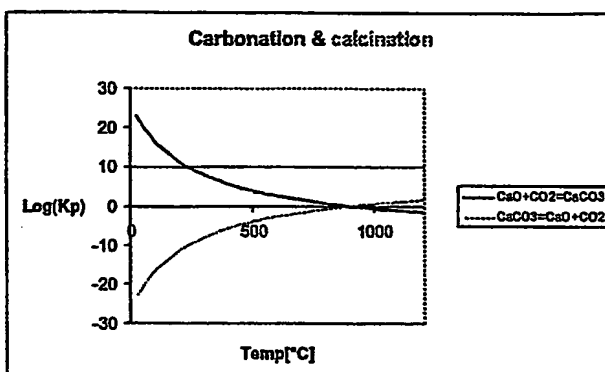


Figure C-8 $\text{Log}(K_p)$ for reactions (C-5) and (C-6) as a function of the temperature

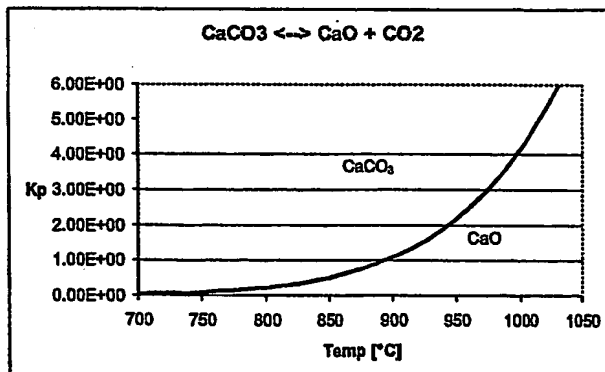



Figure C-9 K_p for reactions (C.5) and (C.6) as a function of the temperature. When CO_2 is the only gas component present, the calcination reaction will occur if $P < p_{\text{CO}_2}$ where P is the pressure inside the calcination reactor and K_p is, in this case, the equilibrium pressure of CO_2 (p_{CO_2})

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| prototech  | FUTURE ENERGY PLANTS Technical Summary Report M1 Appendix C: - NG-WATER REFORMING, CARBONATION and CALCINATION | Doc.no. RP_21205_PR_01 |
| Project: ZEG | | Issue: A Date: 09.09.2002 |
| | | Page: 66 of 73 |

C2.1 Experimental Setup

The experimental set-up is shown in figure C-10 and the explanation follows:

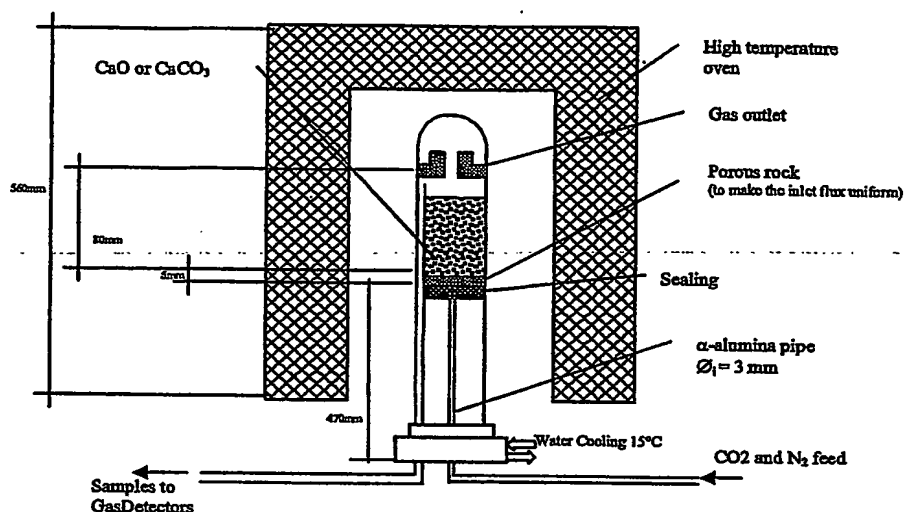


Figure C-10 Experimental set-up.

An alumina tube-continuous-reactor operating at atmospheric pressure in a high temperature-controlled oven has been used.

The feed was consisted of carbon dioxide and nitrogen.

The feed was supplied by means of mass flowmeters.

The nitrogen has been used during the heating up period and the shut down period to create an inert atmosphere and during experiments as a reference.

The sampling has been done after the end of the CaO/CaCO₃ bed.

The instrumental detection has been carried out using IR Gas Detectors.

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C2.2 Experimental results and conclusions

Capture and desorption of CO₂ cyclically using the same starting sample of CaO (converted from sofno lime with a Calcium Hydroxide content >75%) as absorbent led to results showed in figure C-11.

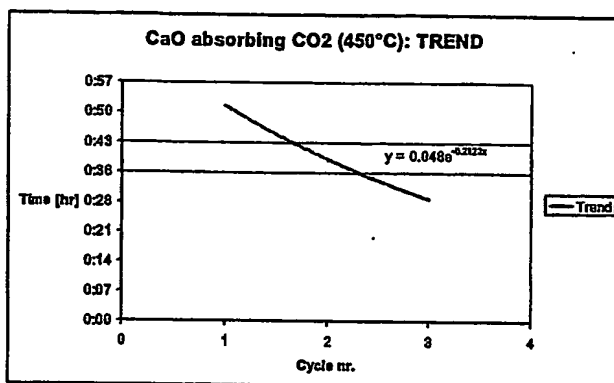


Figure C-11 CaO "deactivation" over cycling. The curve illustrates the reduced absorption capacity of the same CaO sample (at fixed CO₂ flowrate) as a function of the number of absorption/desorption cycles

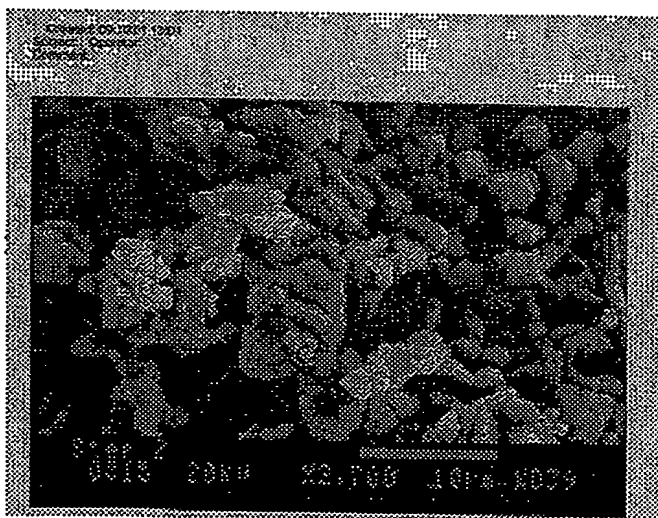


Figure C-12 SEM of a sample of CaO at 1st cycle

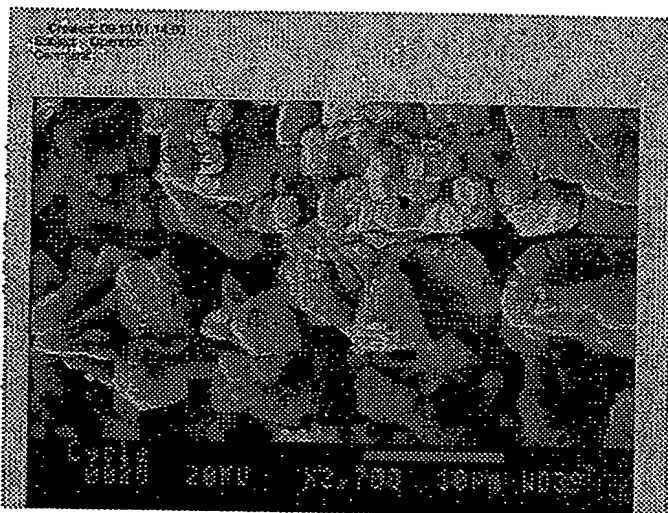


Figure C-13 SEM of a sample of CaO exposed to absorption-desorption cycling; more sharp faces and laminated grains can be noticed if compared with figure C-12

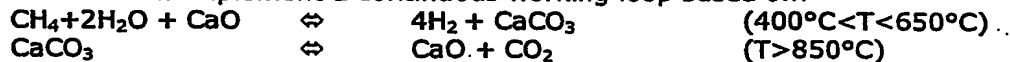
The main conclusions of the study can be summarised as follows:

- There is evidence of sintering or selective growth effect (figure C-13 vs Figure C-12) → the process becomes less efficient with increasing cycling (figure C-11).
- Grinding the material (during the regeneration step) may allow for continuous operation
- Use of a Fluidised Bed (FB) could prevent particles from sintering.

Other experiments [R7] indicate that the absorption capability reaches a limit value after a large number of cycles. The baseline reactor design can still be used without modifications, although absorption/desorption cycling will have to be performed more frequently. Further work will be dedicated to determine and improve the long term metal oxide stability.

C2.3 Discussion and conclusions

Integration of the absorption process with the NG-water reforming is possible over a wide range of operating conditions (including atmospheric pressure coupled with standard (approximately 500°C) reforming temperatures) [R8] and using CaO or calcined dolomite as absorbent [R8]. According to experimental results (see also [R7]) activity loss in the reforming&carbonation process can be attributed to the absorbent. Further work is needed to better identify reaction rates in order to implement a continuous working loop based on:



The feasibility of a batchwise process is experimentally evident [R7, R8] and in the ZEG concept process analysis such a solution has been implemented.

Enthalpies for the reactions are shown in figure C-14. The energy required for the calcination process is then released in the carbonation process that close to balance the energy required by the NG-water reforming process (including the CO shift reaction (1.2)).

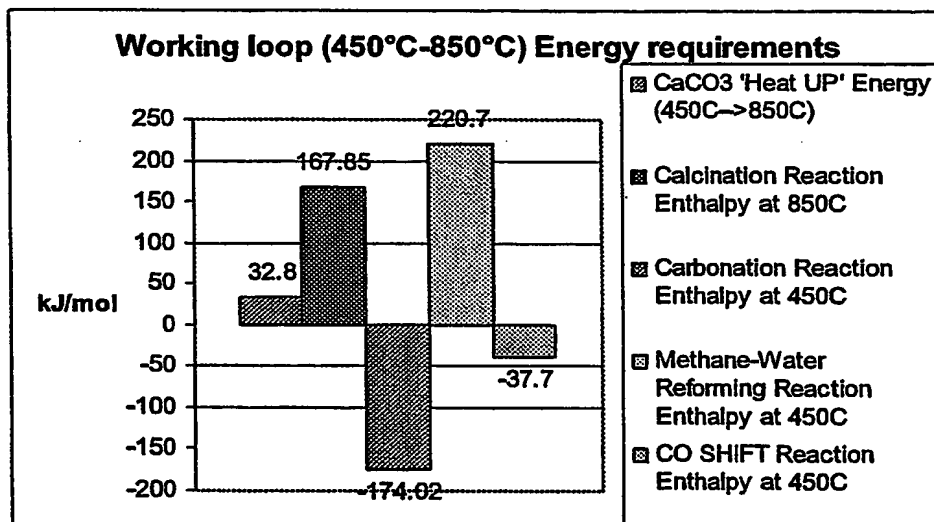



Figure C-14 Reactions enthalpies


| | | |
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| prototech  | FUTURE ENERGY PLANTS Technical Summary Report M1 | Doc.no. RP_21205_PR_01 Issue: A Date: 09.09.2002 |
| Project: ZEG | Appendix D: - MEMBRANE TECHNOLOGY | Page: 70 of 73 |

APPENDIX D: MEMBRANE TECHNOLOGY

The hydrogen flux through a palladium layer can be written as [R9]:

$$\phi = 2.85 \cdot 10^{-7} \exp\left(\frac{-17500}{RT_m}\right) \cdot \left(\frac{p_f^{\frac{1}{2}} - p_p^{\frac{1}{2}}}{t_m}\right) \quad (D.1)$$

where ϕ is the flux [mol/(m²s)]; t_m is effective thickness of the membrane [m]; R is the gas constant [J/(mol*K)]; T_m is the absolute temperature [K] of the membrane; p_f is the feed pressure [Pa]; p_p is the permeate pressure [Pa].

| | | |
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| prototech  | FUTURE ENERGY PLANTS Technical Summary Report M1 | Doc.no. RP_21205_PR_01 Issue: A Date: 09.09.2002 |
| Project: ZEG | Appendix D: - MEMBRANE TECHNOLOGY | Page: 71 of 73 |

According to eq.(D.1) and analysing the data concerning concept ZEG 3 the following is obtained.

| | |
|--|--|
| $dh_{H_2} := 241.827 \frac{\text{kJ}}{\text{mole}}$ | LHV of H ₂ at 25°C |
| $W_{H_2} := 7.5 \text{ MW}$ | H ₂ to filter (LHV basis) |
| $m_{H_2} := \frac{W_{H_2}}{dh_{H_2}}$ | $m_{H_2} = 31.014 \text{ s}^{-1} \cdot \text{mol}$ |
| $R = 8.314 \text{ mol}^{-1} \frac{\text{J}}{\text{K}}$ | GAS CONSTANT |
| $T_m := (450 + 273) \cdot \text{K}$ | Operating Temp |
| $t_m := 4 \cdot \mu\text{m}$ | Membrane Thickness |
| $p_f := 4 \cdot \text{bar}$ | Inlet Pressure (retentate) |
| $p_p := 1 \cdot \text{bar}$ | Outlet Pressure |
| $\phi := 2.85 \cdot 10^{-7} \cdot \exp\left(-\frac{17500 \frac{\text{J}}{\text{mole}}}{R \cdot T_m}\right) \cdot \left[\frac{\left(\frac{p_f}{\text{Pa}}\right)^{0.5} - \left(\frac{p_p}{\text{Pa}}\right)^{0.5}}{\left(\frac{t_m}{\text{m}}\right)} \right] \frac{\text{mole}}{\text{m}^2 \cdot \text{s}}$ | |
| $\phi = 1.226 \frac{\text{mole}}{\text{m}^2 \cdot \text{s}}$ | Flux through the membrane |
| $A_m := \frac{m_{H_2}}{\phi}$ | |
| $A_m = 25.299 \text{ m}^2$ | Required membrane area |

$\phi \propto \frac{1}{t_m}; A_m \propto \frac{1}{\phi} \Rightarrow \text{Volume}_{Pd} \propto t_m^2 \Rightarrow$ the membrane thickness is the key factor for the total material cost.

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APPENDIX E: OXYGEN PUMP

The following calculations led to the power consumption estimation for the oxygen pump required in concepts ZEG 1 and 3.

Solid Oxide Cell physical properties

| | | |
|---|---|--|
| Cathode specific conductivity | $\lambda_k := 12870 \frac{\text{S}}{\text{m}}$ | |
| Electrolyte specific conductivity | $\lambda_e := 18 \frac{\text{S}}{\text{m}}$ | |
| Resistance | $R_o(\lambda, B, T, H) := \frac{H}{\lambda \cdot B \cdot T}$ | |
| Specific resistance | $r(\lambda, B, T, H, A_r) := \frac{H \cdot A_r}{\lambda \cdot B \cdot T}$ | |
| B*T=ConductiveArea, H=ConductiveLength, A_r=ReferenceArea | | |
| | $A_r := 67 \cdot \text{cm}^2$ | |
| Electrolyte resistance | $r_e := r(\lambda_e, 1 \cdot \text{cm}, 1 \cdot \text{cm}, 150 \cdot \mu\text{m}, 1 \cdot \text{cm}^2)$ | |
| | $r_e = 0.083 \cdot \Omega \cdot \text{cm}^2$ | |
| | $\frac{r_e}{A_r} = 1.24378 \cdot 10^{-3} \cdot \Omega$ | |
| Cathode resistance (vertical specific) | $r_{k1} := r(\lambda_k, 1 \cdot \text{cm}, 0.1 \cdot \text{cm}, 35 \cdot \mu\text{m}, 1 \cdot \text{cm}^2)$ | |
| | $r_{k1} = 2.72 \cdot 10^{-4} \cdot \Omega \cdot \text{cm}^2$ | |
| Contact resistance (specific) | $R_c := 0.0005 \Omega$ | |
| | $r_c := R_c \cdot 67 \cdot \text{cm}^2$ | $r_c = 0.034 \cdot \Omega \cdot \text{cm}^2$ |
| Total cell resistance (specific) | $r_{tot} := r_e + 2 \cdot r_{k1} + r_c$ | $r_{tot} = 0.117 \cdot \Omega \cdot \text{cm}^2$ |

Solid Oxide Cell physical properties (continued)

| | | |
|--|---|--|
| Total cell resistance | $R_{tot} := \frac{r_{tot}}{67 \cdot \text{cm}^2}$ | $R_{tot} = 1.752 \cdot 10^{-3} \Omega$ |
| Outer voltage (applied) | $U_o := 0.1 \cdot \text{V}$ | |
| Difference in potential | | |
| $T := 1173 \cdot \text{K}$ | $p_{O2in} := 0.21 \cdot \text{bar}$ | $p_{O2out} := 1 \cdot \text{bar}$ |
| Differential voltage | $U_d := \frac{R \cdot T}{4 \cdot F} \cdot \ln\left(\frac{p_{O2out}}{1 \cdot \text{bar}}\right) - \frac{R \cdot T}{4 \cdot F} \cdot \ln\left(\frac{p_{O2in}}{1 \cdot \text{bar}}\right)$ | |
| | $U_d = 39.437 \cdot \text{mV}$ | |
| Current | $I_{oxy} := \frac{U_o - U_d}{R_{tot}}$ | $I_{oxy} = 34.57 \cdot \text{A}$ |
| Molar flow of Oxygen per "O ₂ cell" | $m_{oxy} := \frac{I_{oxy}}{4 \cdot F}$ | $m_{oxy} = 8.957 \cdot 10^{-5} \frac{\text{mole}}{\text{s}}$ |

Oxygen pump power

| | | |
|---|--|-------------------------------------|
| Power for oxygen pump where N_c is the number of required "O ₂ cells" | $W_{oxy} := U_o \cdot I_{oxy} \cdot N_c$ | $W_{oxy} = 189.076 \cdot \text{kW}$ |
| $P_e =$ gross plant power output (7.5 [MW _{el}]) | | $\frac{W_{oxy}}{P_e} = 0.025$ |
| Thermodynamic compression power: isothermal compression where m_{O_2} is the required molar flow [mol/sec] of oxygen | $W_{td} := m_{O_2} \cdot R \cdot T \cdot \ln\left(\frac{p_{O2out}}{p_{O2in}}\right)$ | $W_{td} = 74.565 \cdot \text{kW}$ |

(When current density approach zero, power approaches theoretic Thermodynamic compression power for isothermal compression.)

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